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(19)(CA) **APPLICATION FOR CANADIAN PATENT** (12)

(54) N-Methylamides, Their Preparation and Intermediates Therefor and Control of Pests Therewith

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(30) (DE) P 42 28 867.3 1992/08/29

(57) 7 Claims

Notice: This application is as filed and may therefore contain an *incomplete specification.*

Canada

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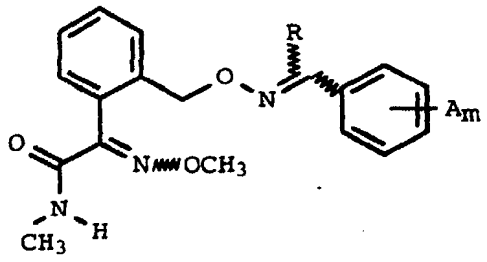
N-Methylamides, their preparation and intermediates therefor and control of pests therewith

ABSTRACT OF THE DISCLOSURE:

N-Methylamides of the formula I,

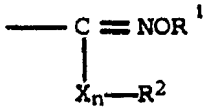


I

where

R is C_1-C_3-alkyl or cyclopropyl,

A is hydrogen, halogen, cyano, nitro, alkyl, cycloalkyl, OR^1, cycloalkyloxy, haloalkyl, haloalkyloxy, alkenyl, alkenyloxy, alkynyl, alkoxyalkyl, cyanoalkyl, nitroalkyl, phenyl, phenoxy, $C(O)R^1$, CO_2R^1, $C(O)NR^1R^2$, $C(S)NR^1R^2$, NR^1R^2, $NR^1C(O)R^2$, $NR^1CO_2R^2$, $OC(O)R^1$, SR^1, $S(O)R^1$, $S(O)_2R^1$, $-C(R^1)=NR^2$, $-N=CR^1R^2$,



where R^1, R^2 and R^3 are hydrogen or C_1-C_6-alkyl and X is S, O or NR^3 and n is 0 or 1 or two of the groups A_m in adjacent positions may together be -CH=CH-CH=CH- and

m is 1, 2 or 3,

and fungicides containing these compounds.

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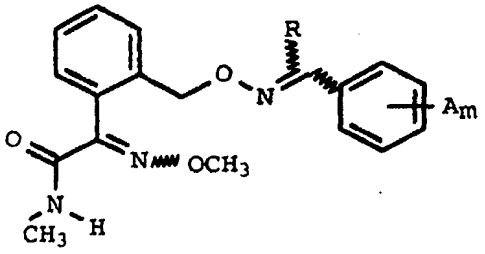
N-Methylamides, their preparation and intermediates therefor and control of pests therewith

The present invention relates to novel N-methylamides, processes and intermediates for their preparation and processes for controlling pests, in particular fungi, insects, nematodes and spider mites, using these compounds.

It is known to use substituted N-methylamides as pesticides (cf. EP 463 488, EP 398 692). However, their action is unsatisfactory. It has surprisingly been found that N-methylamides of the general formula I



I

have an excellent fungicidal, insecticidal, nematicidal and acaricidal action which is better than that of the known N-methylamides.

The fungicidal action is preferred.

The radicals mentioned under the general formula I can have, for example, the following meanings:

R
can be C_1-C_3-alkyl (eg. methyl, ethyl, n- or isopropyl) or cyclopropyl,

A
can be identical or different and is hydrogen, halogen (eg. fluorine, chlorine, bromine or iodine), cyano, nitro, C_1-C_6-alkyl (eg. methyl, ethyl, n- or isopropyl, n-, iso-, sec- or tert-butyl, n-pentyl, neopentyl, tert-pentyl or hexyl),

or C_3-C_6-cycloalkyl (eg. cyclopropyl, cyclobutyl, cyclopentyl or cyclohexyl),

OR^1
(eg. hydroxyl, methoxy, ethoxy, n-propoxy, isopropoxy, n-, iso-, sec- or tert-butoxy, n-pentoxy, tert-pentoxy, neopentoxy or n-hexoxy),

C_3-C_6-cycloalkoxy (eg. cyclopropoxy, cyclobutoxy, cyclopentoxy or cyclohexoxy),

C_1-C_6-haloalkyl (eg. trifluoromethyl, 2-fluoroethyl, 2,2,2-trifluoroethyl, pentafluoroethyl, fluorodichloromethyl, difluorochloromethyl, chloromethyl, dichloromethyl, trichloromethyl, 2-chloroethyl, 2,2,2-trichloroethyl, pentachloroethyl or 3-chloro-2-methylprop-2-yl),

C_1-C_6-haloalkoxy (eg. difluoromethoxy, trifluoromethoxy, pentafluoroethoxy, 1,1,2,2-tetrafluoroethoxy or 2,2,2-trifluoroethoxy),

C_2-C_6-alkenyl (vinyl, 1-propen-1-yl, 2-propenyl, 2-butenyl, 1-methyl-2-propenyl, allyl, but-2-en-2-yl),

C_2-C_6-alkenyloxy (eg. vinyloxy or allyloxy),

C_2-C_6-alkynyl (ethynyl, 1-propynyl or 3-propynyl),

C_1-C_6-alkoxy-C_1-C_6-alkyl (eg. methoxymethyl, ethoxymethyl, dimethoxymethyl, 1,1-dimethoxyeth-1-yl), n- or isopropoxymethyl, n-, iso-, sec- or tert-butoxymethyl, 1-methoxy-2-methylprop-2-yl, 2-methoxyprop-2-yl, 2-ethoxyprop-2-yl, 2-n- or isopropoxyprop-2-yl, 2-n-, iso-, sec- or tert-butoxyprop-2-yl),

cyano-C_1-C_6-alkyl (eg. cyanomethyl, 1-cyano-2-methylprop-2-yl or 2-cyanoethyl),

nitro-C_1-C_6-alkyl (eg. nitromethyl),

phenyl or phenoxy,

$C(O)R^1$ (eg. formyl, acetyl, propionyl, butyryl, isobutyryl or pivaloyl),

CO_2R^1 (eg. hydroxycarbonyl, methoxycarbonyl, ethoxycarbonyl, n- or isopropoxycarbonyl, n-, iso-, sec- and tert-butoxycarbonyl),

$C(O)NR^1R^2$ (eg. aminocarbonyl, N-methylaminocarbonyl, dimethylaminocarbonyl or diethylaminocarbonyl),

$C(S)NR^1R^2$ (eg. aminothiocarbonyl, N-methylaminothiocarbonyl or N,N-dimethylaminothiocarbonyl),

NR^1R^2 (eg. amino, N-methylamino, N-ethylamino, N-isopropylamino, N-n-propylamino, N-n-butylamino, N-sec-butylamino, N-tert-butylamino, N-isobutylamino or N,N-dimethylamino),

$NR^1C(O)R^2$ (eg. N-acylamino, N-methyl-N-acylamino or N-ethyl-N-acylamino),

$NR^1CO_2R^2$ (eg. N-carboxymethylamine or N-methyl-N-carboxymethylamine),

$OC(O)R^1$ (eg. OC(O)methyl, OC(O)ethyl, OC(O)n-propyl, OC(O)i-propyl, OC(O)n-butyl or OC(O)tert-butyl),

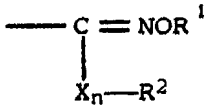
SR^1 (eg. S-H, S-methyl, S-ethyl, S-n-propyl, S-i-propyl, S-n-butyl, S-sec-butyl, S-isobutyl, S-tert-butyl, S-n-pentyl or S-n-hexyl),

$S(O)R^1$ (eg. S(O)-methyl, S(O)-ethyl or S(O)n-propyl,

SO_2R^1 (eg. SO_2-methyl or SO_2-ethyl),

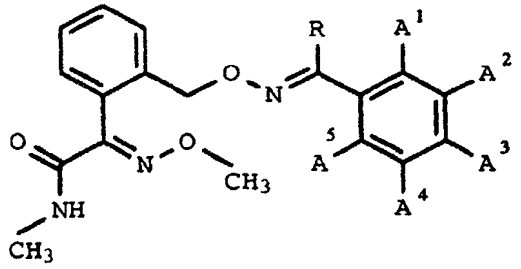
$-C(R^1)=NR^2$ (eg. -CH=NH, -CH=N-methyl, -CH=N-ethyl, $-C(CH_3)=NH$, $-C(CH_3)=NCH_3$),

$-N=CR^1R^2$ (eg. $-N=C(CH_3)_2$, $-N=C(CH_3)$(ethyl)),



(eg. methoxyiminomethyl, ethoxyiminomethyl, n-propoxyiminomethyl, n-butoxyiminomethyl, methoxyimino-1-ethyl, ethoxyimino-1-ethyl, n-propoxyimino-1-ethyl, n-butoxyimino-1-ethyl, i-propoxyimino-1-ethyl, sec-, iso- or tert-butoxyimino-1-ethyl, n-pentoxyimino-1-ethyl, n-hexoxyimino-1-ethyl, -C(OMe)=NOMe, -C(SMe)=NOMe, $-C(NH_2)$=NOMe, $-C(NMe_2)$=NOMe, -C(NHMe)=NOMe, -C(OEt)=NOEt, -C(SMe)=NOEt), with the exception of compounds of the formula I *in which R is methyl and A_m is 2-chloro, 3-chloro, 4-chloro,* 3,5-dichloro, 2,3,4-trichloro, 2-methyl, 4-methyl, 3-bromo, 4-nitro or hydrogen and with the exception of compounds of the formula I in which R is cyclopropyl and A_m is hydrogen, 4-chloro, 4-tert-butyl or 4-methoxy.

The invention also relates to compounds of the formula Ia



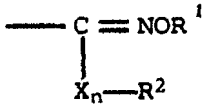
Ia

where

R is methyl, ethyl, n-propyl or isopropyl,

A^1, A^2, A^3, A^4 and A^5 are identical or different and are hydrogen, halogen, cyano, nitro, C_1-C_6-alkyl, C_3-C_6-cycloalkyl, OR^1, C_3-C_6-cycloalkoxy, C_1-C_6-haloalkyl, C_1-C_6-haloalkoxy, C_2-C_6-alkenyl, C_2-C_6-alkenyloxy, C_2-C_6-alkynyl, C_1-C_6-alkoxy-C_1-C_6-alkyl, cyano-C_1-C_6-alkyl, nitro-C_1-C_6-alkyl, phenyl, phenoxy, $C(O)R^1$, CO_2R^1, $C(O)NR^1R^2$, $C(S)NR^1R^2$, NR^1R^2, $NR^1C(O)R^2$, $NR^1CO_2R^2$, $OC(O)R^1$, SR^1, $S(O)R^1$, $S(O)_2R^1$, the groups

$-C(R^1)=NR^2$, $-N=CR^1R^2$,



where the radicals R^1, R^2 and R^3 independently of one another are hydrogen or C_1-C_6-alkyl and X is S, O or NR^3 and n is 0 or 1, or two of the groups A^1 to A^5 in adjacent positions are the group -CH=CH-CH=CH-

with the proviso that

a) two to four of the substituents A^1 to A^5 are hydrogen,

b) two to three of the substituents A^1 to A^5 are hydrogen if one of the substituents A^1 to A^5 is Cl, Br, NO_2 or CH_3,

c) one of the halogen atoms is F or Br if A^2+A^4 or A^1+A^2+A^3 are simultaneously halogen.

Preferred substituents for R are methyl and ethyl, in particular methyl, and for A or A^1 to A^5 are hydrogen, fluorine, chlorine, bromine, methyl, ethyl, isopropyl, tert-butyl, tert-pentyl, cyano, methoxy, ethoxy, isopropoxy, tert-butoxy, tert-pentoxy, me-

thoxymethyl, 2-methoxyprop-2-yl, trifluoromethyl, 1,1,2,2tetrafluoroethyl, 1-chloro-2-methylprop-2-yl and trifluoromethoxy.

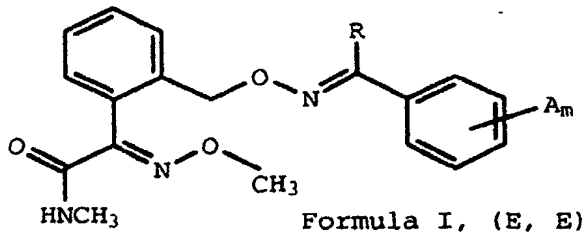
Preferred compounds are those whose substituents A other than hydrogen are in the positions A^2, A^3 or A^4.

Preferred compounds in this case are those which result if the substituents other than hydrogen represent the following substition pattern.

a) A^2 (or A^4) $\neq$ H; all other substituents = H
b) A^3 $\neq$ H; all other substituents = H
c) A^2 and A^3 (or A^3 and A^4) $\neq$ H; all other substituents = H
d) A^2 and A^4 $\neq$ H; all other substituents = H
e) A^2 and A^3 and A^4 $\neq$ H; all other substituents = H
($\neq$ means: other than)

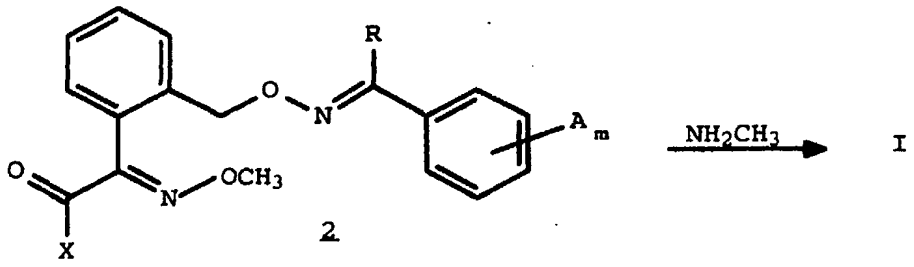
Particularly preferred substitution patterns are those with the substituents A^2, A^3 and A^4 which are given above as preferred.

On account of the C=N double bonds, the novel compounds of the general formula I can be obtained as E/Z isomer mixtures. These can be separated into the individual components in conventional manner, for example by crystallization or chromatography. Both the individual isomeric compounds and their mixtures are covered by the invention and can be used as pesticides. As a rule, however, the compounds according to the invention are mainly obtained in the (E, E) configuration. These isomers are also preferred with respect to their action.



Formula I, (E, E)-configuration

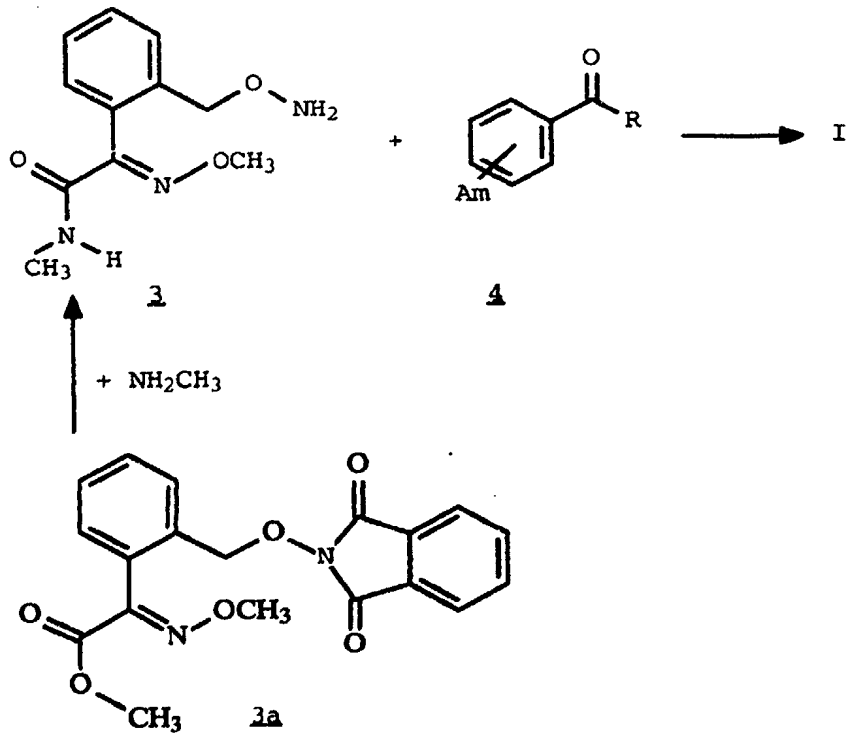
The novel compounds of the formula I can be prepared, for example, as follows:



Starting from the compounds of the formula 2, where X = OH, Cl or O(C_1-C_4-alkyl) [disclosed in EP 463 488, 472 300 and 426 460], the monomethylamides I according to the invention can be obtained analogously to methods known from the literature by reaction with methylamine (cf. Organikum; 16th edition (1985) p. 408 et seq).

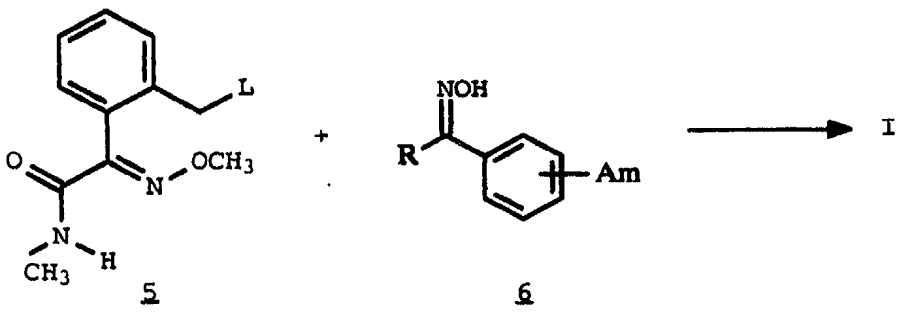
A particularly advantageous method of preparing the compounds I is to react O-benzylhydroxylamine 3 with the phenylketones 4 (cf. D. Otzanak, J. Chem. Soc., Chem. Commun. 1986, 903). The intermediate 3 required for this can surprisingly be prepared particularly smoothly and advantageously in a simple reaction by triple aminolysis of 3a (with methylamine).

This one-step reaction procedure 3a → 3 avoids the conventional more involved and more elaborate two-step procedure, eg. setting free the O-benzylhydroxylamine group from the phthalimide protective group with the aid of hydrazinolysis and beforehand or subsequently converting the methyl ester group to the methylamide. The high yields of this reaction are surprising, although normally the optimum conditions are very different on the one hand for the aminolysis of a methyl ester to the methylamide and on the other hand for setting free an O-benzylhydroxylamine from the corresponding O-benzylhydroxyphthalimide.



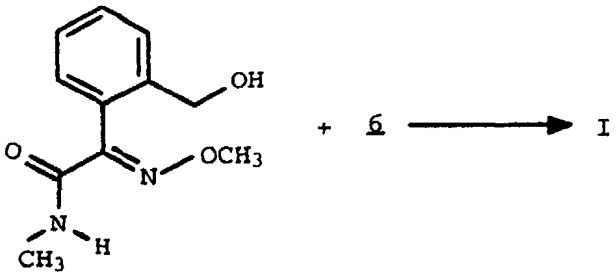
The phenylketones 4 are known or can be prepared analogously to known processes (Houben-Weyl, Vol. 7/2a and 7/3a).

The novel compounds of the formula I are also obtained by reacting a monomethylamide benzyl compound 5, in which L is a leaving group (eg. bromine, chlorine, iodine, mesylate, tosylate or triflate) (cf., eg. EPs 398 692 and 463 488), with an oxime of the formula 6 (cf., eg. Houben-Weyl, Vol. 10/1, p. 1186 et seq).



The oximes 6 are known or can be prepared by known processes (cf. Houben-Weyl, Vol. 10/4; J. Med. Chem. 26, (1983) 1360).

The novel compounds of the formula I can furthermore be prepared by reacting the benzyl alcohol 7 with oximes 6 in conventional manner (cf. J. Jurczak, Synthesis, 1976, 682).



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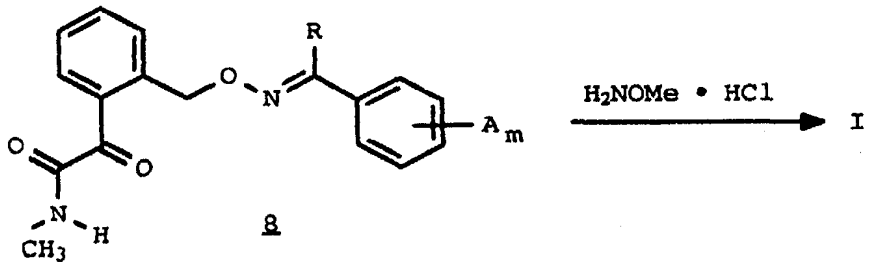
Alternatively, for this purpose the ketoamides 8 can be reacted in conventional manner with methoxylamine (hydrochloride) to give the novel compounds I [cf., eg. DE 3623921).



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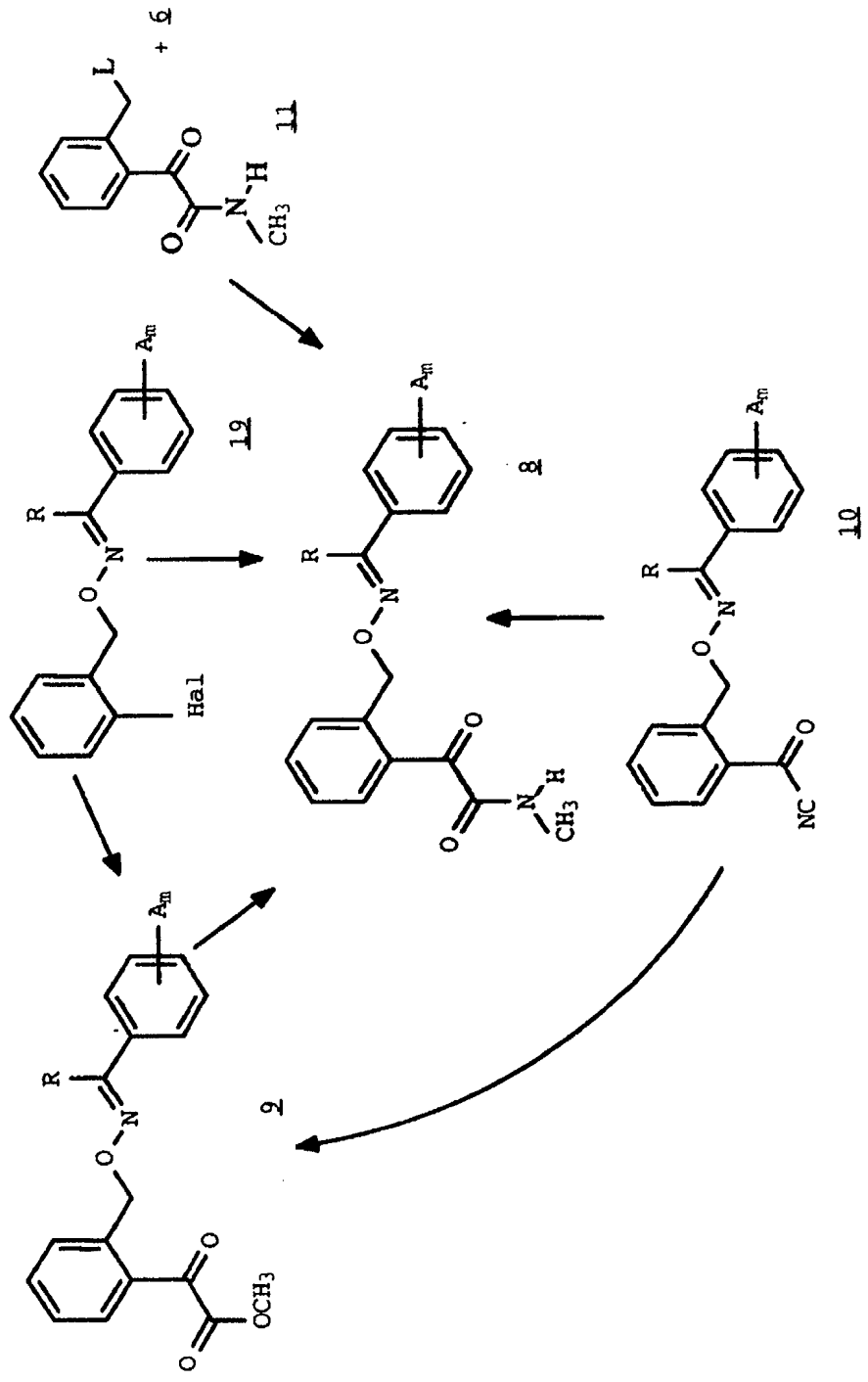
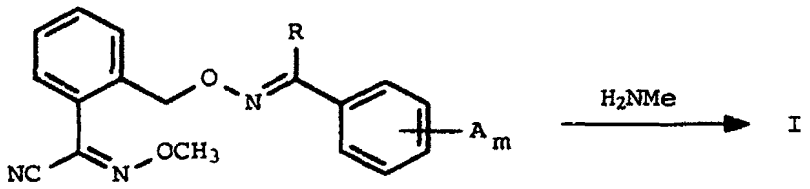
The ketoamides 8 themselves can be obtained, for example, from the ketoesters 9 by aminolysis, the ketoesters 9 themselves being accessible either by Pinner reaction from benzoyl cyanides 10 or by reaction of oximes 6 with benzyl compounds 11.

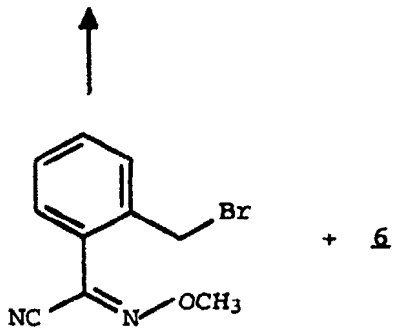

+ 6
11
L
O
O
N
H
CH3
Am
R
N
O
19
Hal
8
10
NC
9
OCH3

Suitable further starting materials for the preparation of the ketoamides 8 or of the ketoesters 9 are aryl halides 19 (Hal = chlorine, bromine or iodine). These can be converted into the corresponding organometal by metallization (eg. with magnesium, methylmagnesium bromide, sodium or n-butyllithium), which can then be reacted with oxalic acid derivatives (e.g., oxalic acid diester, oxalic acid ester N-methylamide) to give the products 8 or 9 (cf., eg. L.M. Weinstock, Synth. Commun. 11, (1981) 943).

The cyanooxime ethers 12 can also be converted with methylamine in conventional manner into the novel compounds of the formula I (cf. EP 468 775).



12

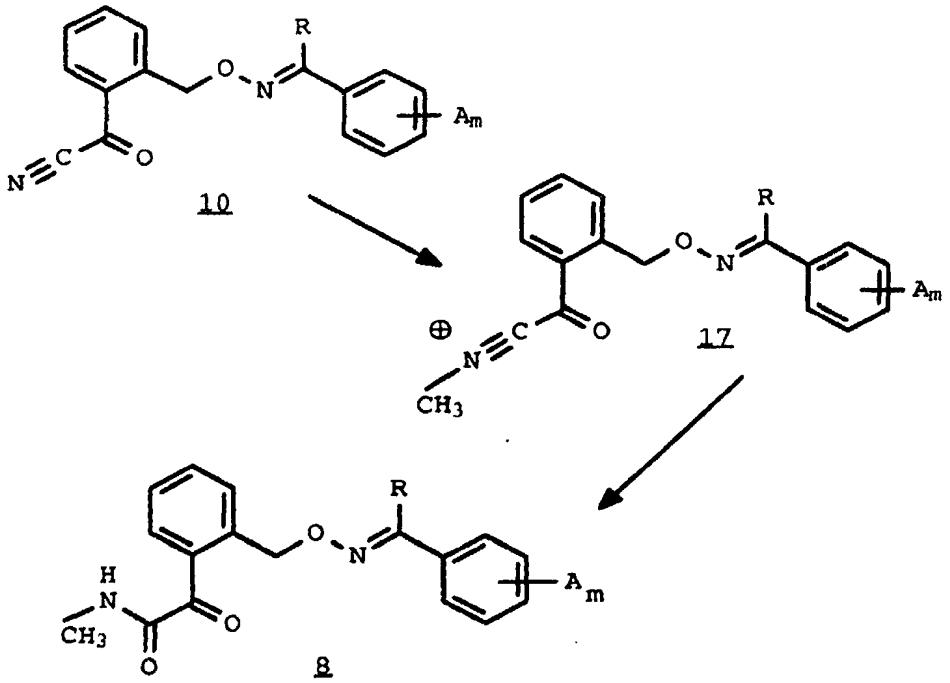


+ 6

13

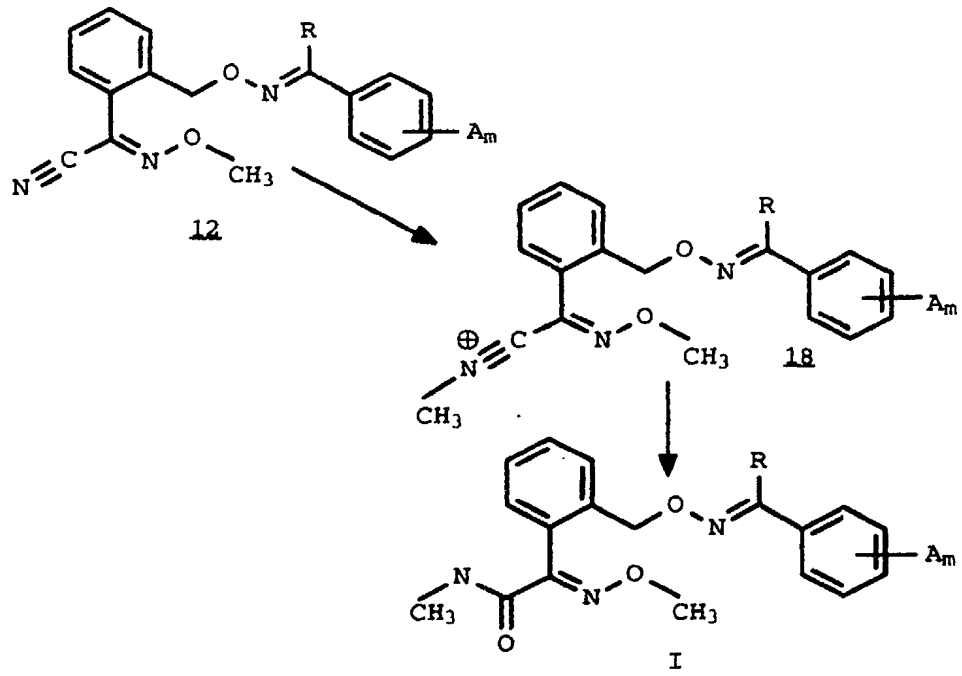
The cyanooxime ethers 12 are obtained, for example, according to known processes by reaction of oximes 6 with benzyl bromide 13.

The ketoamides of the formula 8 can additionally be obtained in a simple manner from nitrilium salts 17 by hydrolysis either in acidic or in alkaline solution (cf. Houben-Weyl, Vol. E5/part 2, p. 1580 et seq).



The nitrilium salts of the formula 17 are easily accessible, for example, from compounds of the formula 10 by reaction with trimethyloxonium salts such as, for example, trimethyloxonium tetrafluoborate or by reaction of 10 with, for example, methyl trifluoromethanesulfonate (cf. Houben-Weyl, Vol. E5/part 2, pp. 1573-6).

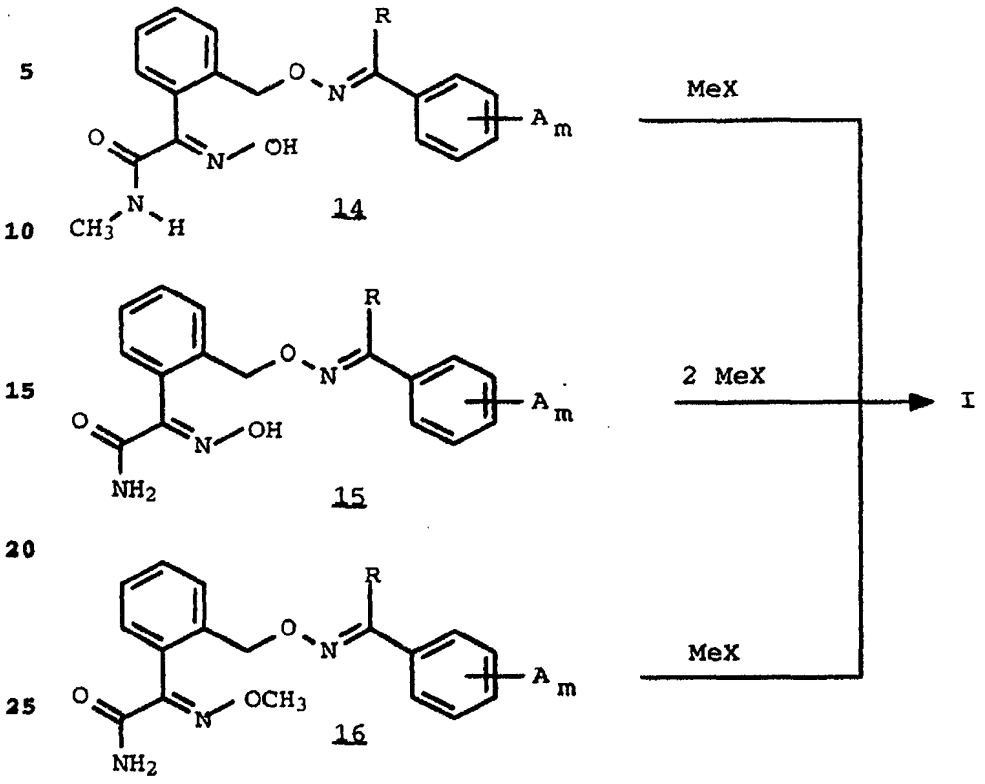
In a corresponding manner, the compounds of the general formula I can also be obtained easily from nitrilium salts of the formula 18 by hydrolysis in acidic or alkaline solution (cf. Houben-Weyl, Vol. E5/part 2, p. 1580 et seq).



12 18 I

The nitrilium salts of the formula 18 in turn are easily accessible, for example, from compounds of the formula 12 by reaction with trimethyloxonium salts such as, for example, trimethyloxonium tetrafluoborate or by reaction of 12 with, for example, methyl trifluoromethanesulfonate (cf. Houben-Weyl, Vol. E5/part 2, pp. 1573-6).

The novel compounds of the formula I can also be prepared by methylation of suitable intermediates such as, for example, 14 - 16.



Alkylation expediently takes place in the presence of a base such as, for example, KOH, K_2CO_3 or triethylamine. Suitable alkylating agents are methyl halides or dimethylsulfate (cf. for example Houben-Weyl, vol. X/1 pp. 1186 ff; or G. L. Isele, Synthesis, 266 (1971)).

The substituents R and A_m shown in the formulae 2 - 19 have the meanings given in claim 1. The substituent L is a leaving group such as, for example, halogen (chlorine, bromine or iodine), tosylate, mesylate or triflate.

The following examples are intended to illustrate the preparation of the novel active compounds and of the novel intermediates:

EXAMPLE 1

Preparation of methyl E-2-methoxyimino-2-(2'-phthalimidooxymethyl)phenylacetate

59 g (0.58 mol) of triethylamine is added dropwise to a solution of 150 g (0.52 mol) of methyl 2-methoxyimino-2-(2'-bromomethyl)phenylacetate and 85 g (0.52 mol) of N-hydroxyphthalimide in 350 ml of N-methylpyrrolidone. The reaction mixture is stirred at 70°C for 2 h and poured into 2 l of ice-water, and the precipitated crystals are filtered off with suction and taken up in methylene chloride. After the organic phase has been washed with water and dried over sodium sulfate, and the solvent has been removed on a rotary evaporator, 168 g (88% yield) of methyl E-2-methoxyimino-2-(2'-phthalimidooxymethyl)phenylacetate is obtained as a pale gray powder of melting point 152 - 153°C.

1H-NMR ($CDCl_3$) δ = 3.81 (s, 3H); 3.95 (s, 3H); 5.05 (s, 2H); 7.12 - 7.81 (m, 8H) ppm.

EXAMPLE 2

Preparation of N-methyl-E-2-methoxyimino-2-(2'-aminooxymethyl)phenylacetamide

168 g (0.46 mol) of methyl E-2-methoxyimino-2-(2'-phthalimidooxymethyl)phenylacetate is treated with 1 l of 40% strength aqueous monomethylamine solution and the mixture is stirred at 40°C for 4 hours. After cooling, it is extracted with methylene chloride, and the organic phase is washed with water, dried over sodium sulfate and concentrated. After crystallization of the residue from methyl tert-butyl ether/n-hexane, 92 g (84% yield) of N-methyl-E-2-methoxyimino-2-(2'-aminooxymethyl)phenylacetamide is obtained as pale yellow crystals of melting point 78 - 81°C. The mother liquor contains further final product.

1H-NMR ($CDCl_3$): δ = 2.91 (d, 3H); 3.94 (s, 3H); 4.58 (s, 2H); 5.28 (s, br, 2H); 6.88 (s, br, 1H); 7.14 - 7.43 (m, 4H) ppm.

EXAMPLE 3

Preparation of N-methyl-2-methoxyimino-2-[2'-(1''-(4'''- tert-butylphenyl)-1''-methyl)iminooxymethyl]phenylacetamide ((E,E)isomer: Tab. 1, No. 118)

A mixture of 4.3 g (18 mmol) of N-methyl-E-2- methoxyimino-2-(2'-aminooxymethyl)phenylacetamide, 3.2 g (18 mmol) of 4-tert-butylacetophenone and 1.0 g of molecular sieve (3 Å) in 10 ml of methanol is treated with 100 mg of p-toluenesulfonic acid monohydrate and stirred at room temperature (20°C) for 16 hours.

After filtering off the molecular sieve, the mixture is concentrated and the residue is crystallized from methyl tert-butyl ether/n-hexane. 4.8 g (68% yield) of the (E,E) isomer of the title compound is thus obtained as colorless crystals of melting point 107 - 110°C.

The stereochemistry of the compound is confirmed by ^{13}C-NMR spectroscopy [$C(CH_3)$=N-methyl group: 13 ppm].

1H-NMR ($CDCl_3$): δ = 1.31 (s, 9H); 2.18 (s, 3H); 2.83 (d, 3H); 3.95 (s, 3H); 5.11 (s, 2H); 6.70 (s, br, 1H); 7.19 - 7.55 (m, 8H) ppm.

20% of [Z(C=NObenzyl), E($C=NOCH_3$)] isomer was also detected in the mother liquor after recrystallization by ^1H-NMR spectroscopy besides the (E,E) isomer (shift of the signal from the benzyl protons by 0.18 ppm to higher field).

EXAMPLE 4

Preparation of (E,E)-N-methyl-2-methoxyimino-2-[2'-(1''-(2'''-naphthyl)-1''-methyl)iminooxymethyl]phenylacetamide (Tab. 1, No. 1)

3.0 g (7.7 mmol) of methyl (E,E)-2-methoxyimino-2-[2'-(1''-(2'''-naphthyl)-1''-methyl)iminooxymethyl]phenylacetate (disclosed in EP 463 488) is treated with 50 ml of 40% strength aqueous monomethylamine solution. The mixture is stirred at 40°C for 6 h and, after cooling, extracted three times with methyl tert-butyl ether, and the organic phase is washed with water, dried over sodium sulfate and concentrated. After triturating the residue with diisopropyl ether, 1.9 g (64% yield) of the title compound is obtained as pale yellow crystals of melting point 92 - 95°C.

1H-NMR ($CDCl_3$):δ = 2.30 (s, 3H); 2.83 (d, 3H); 3.94 (s, 3H); 5.15 (s, 2H); 6.69 (s, br, 1H); 7.14 - 7.93 (m, 11H) ppm.

EXAMPLE 5

Preparation of (E,E)-N-methyl-2-methoxyimino-2-[2'-(1''-(4'''-chlorophenyl)-1''-n-propyl)iminooxymethyl]phenylacetamide (Tab. 2, No. 44).

0.6 g (1.5 mmol) of methyl (E,E)-2-methoxyimino-2-[2'-(1''-(4'''-chlorophenyl)-1''-n-propyl)iminooxymethyl]phenylacetate (disclosed in EP 463 488) is dissolved in 15 ml of tetrahydrofuran, and the solution is treated with 0.5 g of 40% strength aqueous monomethylamine solution. The mixture is stirred at 40 - 45°C for 6 hours and then concentrated, the residue is taken up in methyl tert-butyl ether, and the organic phase is extracted with water, dried over sodium sulfate and concentrated again. After chromatography of the residue on a silica gel column (cyclohexane/methyl tert-butyl ether = 1:1), 0.5 g (83% yield) of the title compound is obtained as a colorless resin.

1H-NMR ($CDCl_3$):δ = 0.94 (t, 3H); 1.54 (m, 2H); 2.70 (t, 2H); 2.89 (d, 3H); 3.97 (s, 3H); 5.11 (s, 2H); 6.74 (s, br, 1H); 7.19 - 7.58 (m, 8H) ppm.

EXAMPLE 6

Preparation of E-2-methoxyimino-2-(2'-methylphenyl)acetonitrile.

188 g (1.68 mol) of dry potassium tert-butoxide is introduced into 2 l of dry toluene and treated rapidly with 200 g (1.53 mol) of ortho-methylbenzyl cyanide and 173 g (1.68 mol) of tert-butyl nitrite in 200 ml of toluene. The mixture warms up to about 70°C, and is stirred for 2 hours and treated with 1 l of methyl tert-butyl ether. The yellow potassium salt is filtered off with suction, washed with methyl tert-butyl ether and dried under reduced pressure at 50°C.

290 g (1.46 mol) of this salt is then introduced into 2.5 l of dry acetone with 20 g (0.15 mol) of potassium carbonate. While 234 g (1.65 mol) of iodomethane are added dropwise, the temperature rises to about 35°C. The mixture is stirred overnight and then partitioned between water and methyl tert-butyl ether. The organic phase is washed with water, dried over Na_2SO_4, concentrated and distilled under reduced pressure (b.p. = 95 - 100°C/

0.3 - 0.1 mm). 229 g (78% total yield) of the title compound are thus obtained as a poorly mobile liquid.

1H-NMR ($CDCl_3$):δ = 2.53 (s, 3H); 4.22 (s, 3H); 7.2 - 7.4 (m, 3H); 7.54 (dd, 1H) ppm.

EXAMPLE 7

Preparation of E-2-methoxyimino-2-(2'-bromomethylphenyl)acetonitrile.

48 g (0.28 mol) of E-2-methoxyimino-2-(2'-methylphenyl)acetonitrile and 54 g (0.30 mol) of N-bromosuccinimide are introduced into 250 ml of carbon tetrachloride. The mixture is illuminated from outside with a mercury vapor lamp (300 W) for 40 min. The succinimide is filtered off with suction and the solution is evaporated under reduced pressure. 62 g (88% yield) of the title compound remain, which is about 80% strength according to the result of the ^{1}H-NMR spectrum.

1H-NMR ($CDCl_3$):δ = 4.27 (s, 3H); 4.80 (s, 2H); 7.4 - 7.5 (m, 3H); 7.69 (dd, 1H) ppm.

EXAMPLE 8

Preparation of 2-methoximino-2-(2'-bromomethylphenyl)acetic acid-N-methylamide

10 g of 2-methoximino-2-[(2-methylphenyloxymethyl)-phenyl]acetic acid-N-methylamide is added to 50 ml of dichloromethane. At 10°C, 9 g of HBr is gassed into this solution. It is then allowed to thaw and is stirred for 3 days at room temperature. 50 ml of dichloromethane is added, and the mixture is washed with 5% strength NaOH and with water, dried and evaporated down. There is obtained 7 g of the title compound as a pale brown solid.

m.p.: 128 - 129°C

^{1}H-NMR ($CDCl_3$ / TMS): S. 2.95 (d, 3H); 3.95 (s, 3H); 4.35 (s, 2H); 6.85 (NH); 7.1-7.5 ppm (m, 4H).

EXAMPLE 9

Preparation of 2-methoximino-2-(2'-chloromethylphenyl)acetic acid-N-methylamid

At 10°C, 18.8 g of a boron trichloride solution (1 molar in n-hexane) is dripped into a solution of 2 g (6.4 mmol) of 2-methoximino-2[2-methylphenyloxymethyl)-phenyl]acetic acid-N-methylamide in 30 ml of dichloromethane, and the mixture is refluxed for 1.5 hours. Subsequently, 8.2 g of methanol is added and the mixture is stirred overnight at room temperature. After washing with 5% strength NaOH and with water, drying and evaporting down, there is obtained 1.2 g (78%) of the title compound as a solid.

^{1}H-NMR ($CDCl_3$ / TMS): δ = 2.96 (s, 3H); 3.96 (s, 3H); 4.46 (s, 2H); 6.86 (NH); 7.13-7.49 ppm (m, 4H).

EXAMPLE 10

Preparation of 2-methoximino-2-[2'-1''-(4'''-methylphenyl)1''-methyl)iminoxymethyl]phenylacetic acid-N-methylamide

A spatula tip of tetra-n-butlyammonium iodide is added to a solution of 2 g (7 mmol) of oxime ether amide bromide (from Example 8) in 20 ml of dichloromethane, and then a solution of 1.2 g (7 mmol) of p-methylacetophenone oxime in 10 ml of dichlormethane. 20 ml of 10% strength NaOH is dripped in and the mixture is refluxed for 6 hours. After phase separation, extraction with dichloromethane, drying and evaporting down, the residue is chromatographed on silica gel with hexane/methyl tert.-butyl ether. 1.1 g (45%) of the title compound remain.

^{1}H-NMR ($CDCl_3$/TMS): δ = 2.18; 2.34 (s, 3H); 2.85 (d, 3H); 3.95 (s, 3H); 5.09 (s, 2H); 6.67 (NH); 7.12-7.50 ppm (m, 8H).

EXAMPLE 11

Preparation of 4-methoxyacetophenone oxime-O-[2-bromo]benzyl ether

Under a nitrogen blanket, a solution of 14 g (85 mmol) of p-methoxyacetophenone oxime is dripped into a suspension of 2.8 g (94 mml) of NaH (80% strength) in 250 ml of DMF, and the solution is stirred for 2 hours at room temperature. A solution of 21.3 g (85 mmol) of o-bromobenzyl chloride in 10 ml of DMF is then dripped in and the mixture is stirred for 1.5 hours. After hydro-

lysis with 10% strength HCl, extraction with methyl tert-butyl ether, drying and evaporating down, 27.7 g (98%) of the title compound remain as a yellowish-brown oil.

^{1}H-NMR ($CDCl_3$ / TMS): δ = 2.21; 3.85 (s, 3H); 5.34 (s, 2H); 6.91-7.66 ppm (m, 8H).

EXAMPLE 12

Preparation of 2-methoximino-2-[2'-(1''-(4'''-methoxyphenyl)1''-methyl)iminooxymethyl]phenylacetic acid-N-methylamide

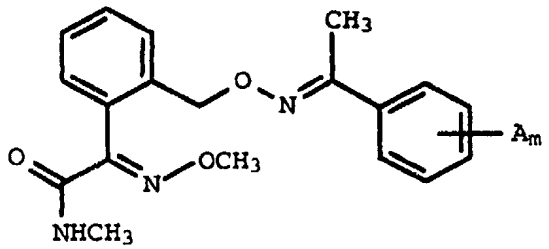
At -78°C and under a nitrogen blanket, 6.6 g (15 mmol) of n-butyllithium (15% strength solution in hexame) is added to a solution of 5 g (15 mmol) of bromide (from Example 11) in 40 ml of THF, and 4.4 g (30 mmol) of diethyl oxalate is run in immediately afterwards. After thawing, water is added, followed by extraction with methyl tert-butyl ether, drying and evaporating down. The residue is chromatographed on silica gel using hexane/methyl tert.-butyl ether. There is obtained 2.7 g (51%) of the corresponding α-keto ester as an oil.

50 ml of a 40% strength n-methylamine solution is added to a solution of 400 mg of this keto ester in 10 ml of THF, and the mixture is heated for 1 hour at 50°C. 2 g of methoxyamine hydrochloride is added and the mixture is again heated for 1 hour at 50°C. After cooling, extraction with dichloromethane, drying and evaporating down, 300 mg of the title compound remains (Table 1, No. 219).

^{1}H-NMR ($CDCl_3$ / TMS): δ = 2.15; 3.13; 3.77 (s, 3H); 2.86 (d, 3H); 5.06 (s, 2H); 6.84-7.56 ppm (m, 8H).

Further compounds of the formula I according to the invention are shown in Tables 1 and 2.

Table 1



No.	A_m	Data
1*		
2+		
3	2F	m.p.: 43 - 47°C
4	3F	oil; IR (film): 3350, 2945, 1671, 1526, 1037, 979
5	4F	m.p.: 60 - 63°C
6	$2,3-F_2$	m.p.: 73 - 75°C
7	$2,4-F_2$	
8	$2,5-F_2$	m.p.: 78 - 80°C
9	$2,6-F_2$	m.p.: 123 - 125°C
10	$3,4-F_2$	m.p.: 103 - 104°C
11	$3,5-F_2$	m.p.: 98 - 100°C
12	$2,3,4-F_3$	
13	$2,3,5-F_3$	
14	$2,4,5-F_3$	m.p.: 67 - 70°C
15	$3,4,5-F_3$	
16	$2,3-Cl_2$	m.p.: 92 - 94°C
17	$2,4-Cl_2$	m.p.: 97 - 99°C
18	$2,5-Cl_2$	m.p.: 118 - 120°C
19	$3,4-Cl_2$	m.p.: 98 - 101°C
20	$3-F,4-OCH_3$	
21	$2,3,5-Cl_3$	
22	$2,4,5-Cl_3$	
23	$3,4,5-Cl_3$	m.p.: 84 - 88°C
24	2-Br	m.p.: 83 - 86°C

No.	A_m	Data
25	4-Br	m.p.: 94 - 96°C
26	2,3-Br_2	
27	2,4-Br_2	
28	2,5-Br_2	
29	3,4-Br_2	
30	3,5-Br_2	m.p.: 120 - 122°C
31	2,3,4-Br_3	
32	2,3,5-Br_3	
33	2,4,5-Br_3	
34	3,4,5-Br_3	
35	2-I	
36	3-I	
37	4-I	
38	2,3-I_2	
39	2,4-I_2	
40	2,5-I_2	
41	3,4-I_2	
42	3,5-I_2	
43	2-F, 3-Cl	
44	2-F, 4-Cl	
45	2-F, 5-Cl	
46	2-F, 3-Br	
47	2-F, 4-Br	
48	2-F, 5-Br	
49	2-Cl, 3-Br	
50	2-Cl, 4-Br	
51	2-Cl, 5-Br	
52	3-F, 4-Cl	
53	3-F, 5-Cl	
54	3-F, 6-Cl	
55	3-F, 4-Br	
56	3-F, 5-Br	
57	3-F, 6-Br	
58	3-Cl, 4-Br	
59	3-Cl, 5-Br	
60	3-Cl, 6-Br	
61	4-F, 5-Cl	m.p.: 150 - 154°C

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No.	A_m	Data
62	4-F, 6-Cl	
63	4-F, 5-Br	m.p.: 80 - 82°C
64	4-F, 6-Br	
65	4-Cl, 5-Br	
66	4-Cl, 6-Br	
67	5-F, 6-Cl	
68	5-F, 6-Br	
69	5-Cl, 6-Br	
70	3-Cl, 4-F, 5-Cl	
71	3-Br, 4-Cl, 5-Br	
72	3-Cl, 4-Br, 5-Cl	
73	2-CN	
74	3-CN	m.p.: 121 - 125°C
75	4-CN	
76	2-NO_2	m.p.: 97 - 99°C
77	3-NO_2	m.p.: 101 - 103°C
78	3-CH_3	oil; IR (film): 3350, 2938, 1671, 1526, 1093, 1037, 979, 787, 697
79	2,3-$(CH_3)_2$	
80	2,4-$(CH_3)_2$	m.p.: 107 - 110°C
81	2,5-$(CH_3)_2$	m.p.: 85 - 86°C
82	2,6-$(CH_3)_2$	
83	3,4-$(CH_3)_2$	
84	3,5-$(CH_3)_2$	m.p.: 88 - 90°C
85	2,3,4-$(CH_3)_3$	
86	2,3,5-$(CH_3)_3$	
87	2,3,6-$(CH_3)_3$	
88	2,4,5-$(CH_3)_3$	
89	2,4,6-$(CH_3)_3$	
90	3,4,5-$(CH_3)_3$	
91	2-C_2H_5	
92	3-C_2H_5	

No.	A_m	Data
93	4-C_2H_5	m.p.: 87 - 89°C
94	2,3-(C_2H_5)$_2$	
95	2,4-(C_2H_5)$_2$	
96	2,5-(C_2H_5)$_2$	
97	3,4-(C_2H_5)$_2$	
98	3,5-(C_2H_5)$_2$	m.p.: 61 - 64°C
99	3,4,5-(C_2H_5)$_3$	
100	3-n-C_3H_7	
101	4-n-C_3H_7	m.p.: 78 - 80°C
101	3,4-(n-C_3H_7)$_2$	
102	3,5-(n-C_3H_7)$_2$	
103	3-i-C_3H_7	
104	4-i-C_3H_7	m.p.: 93 - 95°C
105	3,4-(i-C_3H_7)$_2$	
106	3,5-(i-C_3H_7)$_2$	
107	3-cyclopropyl	
108	4-cyclopropyl	
109	3-n-C_4H_9	
110	4-n-C_4H_9	
111	3,4-(n-C_4H_9)$_2$	
112	3,5-(n-C_4H_9)$_2$	
113	3-s-C_4H_9	
114	4-s-C_4H_9	
115	3-i-C_4H_9	
116	4-i-C_4H_9	
117	3-t-C_4H_9	
118	4-t-C_4H_9	m.p.: 107 - 110°C
119	3,4-(t-C_4H_9)$_2$	
120	3,5-(t-C_4H_9)$_2$	
121	3-n-C_5H_{11}	
122	4-n-C_5H_{11}	
123	3-n-C_6H_{13}	
124	4-n-C_6H_{13}	
125	3-Cyclohexyl	
126	4-Cyclohexyl	m.p.: 110 - 112°C

No.	A_m	Data
127	3-Phenyl	m.p.: 70 - 72°C
128	4-Phenyl	m.p.: 138 - 140°C
129	3-Vinyl	
130	4-Vinyl	
131	3-Allyl	
132	4-Allyl	
133	3-Propargyl	
134	4-Propargyl	
135	3-(Propen-2-yl)	
136	4-(Propen-2-yl)	
137	3-(But-2-en-2-yl)	
138	4-(But-2-en-2-yl)	
139	3-t-C_5H_{11}	
140	4-t-C_5H_{11}	
141	2-CH_3, 3-C_2H_5	
142	2-CH_3, 4-C_2H_5	
143	2-CH_3, 5-C_2H_5	
144	2-CH_3, 3-n-C_3H_7	
145	2-CH_3, 4-n-C_3H_7	
146	2-CH_3, 5-n-C_3H_7	
147	2-CH_3, 3-i-C_3H_7	
148	2-CH_3, 4-i-C_3H_7	
149	2-CH_3, 5-i-C_3H_7	
150	2-CH_3, 3-n-C_4H_9	
151	2-CH_3, 4-n-C_4H_9	
152	2-CH_3, 5-n-C_4H_9	
153	2-CH_3, 3-s-C_4H_9	
154	2-CH_3, 4-s-C_4H_9	
155	2-CH_3, 5-s-C_4H_9	
156	2-CH_3, 3-i-C_4H_9	
157	2-CH_3, 4-i-C_4H_9	
158	2-CH_3, 5-i-C_4H_9	
159	2-CH_3, 3-t-C_4H_9	
160	2-CH_3, 4-t-C_4H_9	
161	2-CH_3, 5-t-C_4H_9	
162	2-CH_3, 3-Cyclohexyl	
163	2-CH_3, 4-Cyclohexyl	
164	2-CH_3, 5-Cyclohexyl	

No.	A_m	Data
165	2-CH_3, 3-Phenyl	
166	2-CH_3, 4-Phenyl	
167	2-CH_3, 5-Phenyl	
168	2-CH_3, 4-Vinyl	
169	2-CH_3, 4-Allyl	
170	2-CH_3, 4-Propargyl	
171	2-CH_3, 4-(Propen-2-yl)	
172	2-CH_3, 4-(But-2-en-2-yl)	
173	3-CH_3, 4-C_2H_5	
174	3-CH_3, 5-C_2H_5	
175	3-CH_3, 4-n-C_3H_7	
176	3-CH_3, 5-n-C_3H_7	
177	3-CH_3, 4-i-C_3H_7	
178	3-CH_3, 5-i-C_3H_7	
179	3-CH_3, 4-Cyclopropyl	
180	3-CH_3, 4-n-C_4H_9	
181	3-CH_3, 5-n-C_4H_9	
182	3-CH_3, 4-s-C_4H_9	
183	3-CH_3, 5-s-C_4H_9	
184	3-CH_3, 4-i-C_4H_9	
185	3-CH_3, 5-i-C_4H_9	
186	3-CH_3, 4-t-C_4H_9	
187	3-CH_3, 5-t-C_4H_9	
188	3-CH_3, 4-Cyclohexyl	
189	3-CH_3, 5-Cyclohexyl	
190	3-CH_3, 4-Phenyl	
191	3-CH_3, 5-Phenyl	
192	3-CH_3, 4-Vinyl	
193	3-CH_3, 4-Allyl	
194	3-CH_3, 4-Propargyl	
195	3-CH_3, 4-(Propen-2-yl)	
196	3-CH_3, 4-(But-2-en-2-yl)	
197	4-CH_3, 5-C_2H_5	
198	4-CH_3, 5-n-C_3H_7	
199	4-CH_3, 5-i-C_3H_7	
200	4-CH_3, 5-Cyclopropyl	
201	4-CH_3, 5-n-C_4H_9	
202	4-CH_3, 5-s-C_4H_9	
203	4-CH_3, 5-i-C_4H_9	

No.	A_m	Data
204	4-CH_3, 5-t-C_4H_9	
205	4-CH_3, 5-Cyclohexyl	
206	4-CH_3, 5-Phenyl	
207	4-CH_3, 5-Vinyl	
208	4-CH_3, 5-Allyl	
209	4-CH_3, 5-Propargyl	
210	4-CH_3, 5-(Propen-2-yl)	
211	4-CH_3, 5-(But-2-en-2-yl)	
212	3-CH_3, 4-t-C_4H_9, 5-CH_3	
213	3-t-C_4H_9, 4-CH_3, 5-t-C_4H_9	
214	2-OH	m.p.: 108 - 110°C
215	3-OH	m.p.: 123 - 125°C
216	4-OH	m.p.: 170 - 171°C
217	2-OCH_3	m.p.: 79 - 81°C
218	3-OCH_3	m.p.: 93 - 95°C
219	4-OCH_3	m.p.: 65 - 68°C
220	2,3-$(OCH_3)_2$	
221	2,4-$(OCH_3)_2$	m.p.: 113 - 114°C
222	2,5-$(OCH_3)_2$	m.p.: 98 - 101°C
223	3,4-$(OCH_3)_2$	m.p.: 132 - 135°C
224	3,5-$(OCH_3)_2$	m.p.: 97 - 99°C
225	3,4,5-$(OCH_3)_3$	m.p.: 118 - 120°C
226	2,3,4-$(OCH_3)_3$	oil; IR (film): 3370, 2945, 1675, 1496, 1463, 1412, 1094, 1037
227	3-OC_2H_5	oil; IR (film): 3350, 2936, 1671, 1526, 1223, 1038, 980
228	4-OC_2H_5	m.p.: 106 - 108°C
229	3,4-$(OC_2H_5)_2$	
230	3,5-$(OC_2H_5)_2$	

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No.	A_m	Data
231	3-O-n-C_3H_7	m.p.: 106 - 109°C
232	4-O-n-C_3H_7	m.p.: 99 - 102°C
233	3,4-(O-n-C_3H_7)$_2$	
234	3,5-(O-n-C_3H_7)$_2$	
235	3-O-i-C_3H_7	oil; IR (film): 3350, 2975, 1672, 152, 1223, 1037, 979
236	4-O-i-C_3H_7	oil; IR (film): 3396, 2980, 1659, 1513, 1252, 1047
237	3,4-(O-i-C_3H_7)$_2$	
238	3,5-(O-i-C_3H_7)$_2$	
239	3-O-Cyclopropyl	
240	4-O-Cyclopropyl	
241	3,4-(O-Cyclopropyl)$_2$	
242	3,5-(O-Cyclopropyl)$_2$	
243	3-O-n-C_4H_9	oil; IR (film): 3360, 2958, 2935, 1671, 1526, 1224, 1037, 979
244	4-O-n-C_4H_9	m.p: 112 - 115°C
245	3,4-(O-n-C_4H_9)$_2$	
246	3,5-(O-n-C_4H_9)$_2$	
247	3-O-s-C_4H_9	
248	4-O-s-C_4H_9	m.p: 95 - 97°C
249	3,4-(O-s-C_4H_9)$_2$	
250	3,5-(O-s-C_4H_9)$_2$	
251	3-O-i-C_4H_9	oil; IR (film): 3350, 2958, 2936, 1672, 1225, 1038, 979
252	4-O-i-C_4H_9	oil; IR (film): 3350, 2959, 2936, 1672, 1514, 1248, 1036
253	3,4-(O-i-C_4H_9)$_2$	
254	3,5-(O-i-C_4H_9)$_2$	
255	3-O-t-C_4H_9	m.p: 66 - 67°C
256	4-O-t-C_4H_9	m.p: 51 - 55°C
257	3,4-(O-t-C_4H_9)$_2$	
258	3,5-(O-t-C_4H_9)$_2$	

No.	A_m	Data
259	3-O-n-C_5H_{11}	oil; IR (film): 3360, 2955, 2935. 1672, 1526, 1226, 1037, 980
260	4-O-n-C_5H_{11}	m.p: 60 - 65°C
261	3,4-(O-n-C_5H_{11})$_2$	
262	3,5-(O-n-C_5H_{11})$_2$	
263	3-O-n-C_6H_{13}	oil; IR (film): 3350, 2933, 1670, 1524, 1223, 1037, 979
264	4-O-n-C_6H_{13}	m.p.: 74 - 76°C
265	3,4-(O-n-C_6H_{13})$_2$	
266	3,5-(O-n-C_6H_{13})$_2$	
267	3-O-Cyclohexyl	
268	4-O-Cyclohexyl	
269	3,4-(O-Cyclohexyl)$_2$	
270	3,5-(O-Cyclohexyl)$_2$	
271	3-OC_6H_5	
272	4-OC_6H_5	m.p.: 118 - 120°C
273	3,4-(OC_6H_5)$_2$	
274	3,5-(OC_6H_5)$_2$	
275	3-O-Allyl	oil; IR (film): 3340, 2942, 1669, 1523, 1225, 1037, 980
276	4-O-Allyl	m.p.: 94 - 97°C
277	3,4-(O-Allyl)$_2$	
278	3,5-(O-Allyl)$_2$	
279	3-O-t-C_5H_{11}	
280	4-O-t-C_5H_{11}	
281	2-CF_3	
282	3-CF_3	oil; IR (film): 3345, 2940, 1672, 1341, 1276, 1167, 1126, 1038
283	4-CF_3	m.p.: 83 - 85°C
284	2,3-(CF_3)$_2$	
285	2,4-(CF_3)$_2$	

No.	A_m	Data
286	2,5-$(CF_3)_2$	
287	3,4-$(CF_3)_2$	
288	3,5-$(CF_3)_2$	m.p.: 138 - 140°C
289	3,4,5-$(CF_3)_3$	
290	2-CF_2Cl	
291	3-CF_2Cl	
292	4-CF_2Cl	
293	3-$CFCl_2$	
294	4-$CFCl_2$	
295	3-CCl_3	
296	4-CCl_3	
297	3,4-$(CCl_3)_2$	
298	3,5-$(CCl_3)_2$	
299	3-CH_2CH_2F	
300	4-CH_2CH_2F	
301	3-CH_2CF_3	
302	4-CH_2CF_3	
303	3-C_2F_5	
304	4-C_2F_5	
305	3-$CHCl_2$	
306	4-$CHCl_2$	
307	3-CH_2Cl	
308	4-CH_2Cl	
309	3,4-$(CH_2Cl)_2$	
310	3,5-$(CH_2Cl)_2$	
311	3-CF_2CHF_2	
312	4-CF_2CHF_2	
313	3-CH_2CCl_3	
314	4-CH_2CCl_3	
315	3-C_2Cl_5	
316	4-C_2Cl_5	
317	3-$C(CH_3)_2CH_2Cl$	
318	4-$C(CH_3)_2CH_2Cl$	
319	3-CH_2CH_2Cl	
320	4-CH_2CH_2Cl	
321	2-OCF_3	
322	3-OCF_3	
323	4-OCF_3	m.p.: 72 - 74°C

No.	A_m	Data
324	2,3-$(OCF_3)_2$	
325	2,4-$(OCF_3)_2$	
326	2,5-$(OCF_3)_2$	
327	3,4-$(OCF_3)_2$	
328	3,5-$(OCF_3)_2$	
329	3-$OCHF_2$	
330	4-$OCHF_2$	m.p.: 78 - 80°C
331	3-$OCCl_3$	
332	4-$OCCl_3$	
333	3-OCH_2CF_3	
334	4-OCH_2CF_3	
335	3,4-$(OCH_2CF_3)_2$	
336	3,5-$(OCH_2CF_3)_2$	
337	3-OC_2F_5	
338	4-OC_2F_5	
339	3,4-$(OC_2F_5)_2$	
340	3,5-$(OC_2F_5)_2$	
341	3-OCF_2CHF_2	m.p.: 67 - 68°C
342	4-OCF_2CHF_2	m.p.: 75 - 77°C
343	3,4-$(OCF_2CHF_2)_2$	
344	3,5-$(OCF_2CHF_2)_2$	
345	3-CF_3, 4-OCF_3	
346	3-CF_3, 5-OCF_3	
347	4-CF_3, 5-OCF_3	
348	3-CH_2OCH_3	
349	4-CH_2OCH_3	
350	3-$CH_2O(t\text{-}C_4H_9)$	
351	4-$CH_2O(t\text{-}C_4H_9)$	
352	3-$C(CH_3)_2CH_2OCH_3$	
353	4-$C(CH_3)_2CH_2OCH_3$	
354	3-$C(CH_3)_2OCH_3$	
355	4-$C(CH_3)_2OCH_3$	
356	3-$C(CH_3)(OCH_3)_2$	
357	4-$C(CH_3)(OCH_3)_2$	
358	3-$CH(OCH_3)_2$	
359	4-$CH(OCH_3)_2$	
360	3-$CH_2(CN)$	

No.	A_m	Data
361	4-$CH_2(CN)$	
362	3-$C(CH_3)_2CH_2CN$	
363	4-$C(CH_3)_2CH_2CN$	
364	3-$CH_2(NO_2)$	
365	4-$CH_2(NO_2)$	
366	3-CHO	
367	4-CHO	
368	3-CO-CH_3	
369	4-CO-CH_3	
370	3-CO-C_2H_5	
371	4-CO-C_2H_5	
372	2-CO_2H	
373	3-CO_2H	
374	4-CO_2H	m.p.: 188 - 190°C
375	3-CO_2CH_3	
376	4-CO_2CH_3	
377	3-$CO_2C_2H_5$	
378	4-$CO_2C_2H_5$	m.p.: 69 - 72°C
379	3-CO_2-n-C_3H_7	
380	4-CO_2-n-C_3H_7	
381	3-CO_2-i-C_3H_7	
382	4-CO_2-i-C_3H_7	
383	3-CO_2-n-C_4H_9	
384	4-CO_2-n-C_4H_9	
385	3-CO_2-s-C_4H_9	
386	4-CO_2-s-C_4H_9	
387	3-CO_2-i-C_4H_9	
388	4-CO_2-i-C_4H_9	
389	3-CO_2-t-C_4H_9	
390	4-CO_2-t-C_4H_9	
391	3-CO_2-n-C_5H_{11}	
392	4-CO_2-n-C_5H_{11}	
393	3-CO_2-n-C_6H_{13}	
394	4-CO_2-n-C_6H_{13}	
395	2-C(=O)-NH_2	
396	3-C(=O)-NH_2	
397	4-C(=O)-NH_2	
398	3-C(=O)-$NHCH_3$	

No.	A_m	Data
399	4-C(=O)-NHCH$_3$	
400	3-C(=O)-N(CH$_3$)$_2$	
401	4-C(=O)-N(CH$_3$)$_2$	
402	3-C(=O)-N(C$_2$H$_5$)$_2$	
403	4-C(=O)-N(C$_2$H$_5$)$_2$	
404	2-C(=S)-NH$_2$	
405	3-C(=S)-NH$_2$	m.p.: 174 - 175°C
406	4-C(=S)-NH$_2$	
407	2-NH$_2$	
408	3-NH$_2$	m.p.: 92 - 94°C
409	4-NH$_2$	131 - 133°C
410	3-NCH$_3$	
411	4-NCH$_3$	
412	3-N(CH$_3$)$_2$	
413	4-N(CH$_3$)$_2$	m.p.: 98 - 100°C
414	3-NHC(=O)CH$_3$	
415	4-NHC(=O)CH$_3$	m.p.: 165 - 167°C
416	3-N(CH$_3$)C(=O)CH$_3$	
417	4-N(CH$_3$)C(=O)CH$_3$	
418	3-NHCO$_2$CH$_3$	
419	4-NHCO$_2$CH$_3$	
420	2-CH$_3$,4-Cyclopropyl	
421	3-N(CH$_3$)CO$_2$CH$_3$	
422	4-N(CH$_3$)CO$_2$CH$_3$	
423	3-O-C(=O)CH$_3$	m.p.: 108 - 110°C
424	4-O-C(=O)CH$_3$	oil; IR (film): 3360, 2942, 1754, 1671, 1199, 1036
425	3,4-[O-C(=O)CH$_3$)$_2$	
426	3,5-[O-C(=O)CH$_3$)$_2$	
427	3-O-C(=O)C$_2$H$_5$	
428	4-O-C(=O)C$_2$H$_5$	
429	3-O-C(=O)(i-C$_3$H$_7$)	
430	4-O-C(=O)(i-C$_3$H$_7$)	
431	3-O-C(=O)(t-C$_4$H$_9$)	
432	4-O-C(=O)(t-C$_4$H$_9$)	

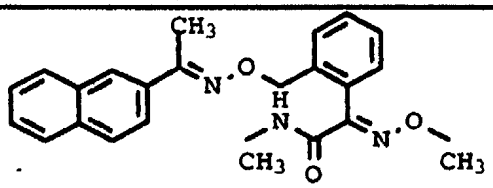
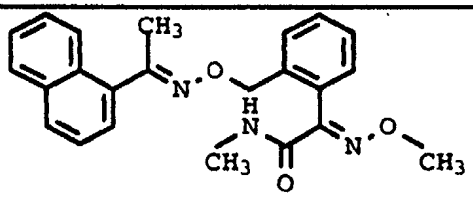
No.	A_m	Data
433	3-SH	
434	4-SH	
435	3-SCH_3	
436	4-SCH_3	m.p.: 97 - 100°C
437	3-SC_2H_5	
438	4-SC_2H_5	
439	3-S-i-C_3H_7	
440	4-S-i-C_3H_7	
441	3-S-t-C_4H_9	
442	4-S-t-C_4H_9	
443	3-S(=O)CH_3	
444	4-S(=O)CH_3	
445	3-SO_2CH_3	
446	4-SO_2CH_3	
447	3-CH=NH	
448	4-CH=NH	
449	3-CH=NCH_3	
450	4-CH=NCH_3	
451	3-C(CH_3)=NH	
452	4-C(CH_3)=NH	
453	3-C(CH_3)=NCH_3	
454	4-C(CH_3)=NCH_3	
455	3-N=C(CH_3)$_2$	
456	4-N=C(CH_3)$_2$	
457	3-C(SCH_3)=NOCH_3	
458	4-C(SCH_3)=NOCH_3	
458	3-C(OCH_3)=NOCH_3	
460	4-C(OCH_3)=NOCH_3	
461	3-C(NH_2)=NOCH_3	
462	4-C(NH_2)=NOCH_3	
463	3-C(NCH_3)=NOCH_3	
464	4-C(NCH_3)=NOCH_3	
465	3-CH=NOCH_3	
466	4-CH=NOCH_3	
467	3-C(CH_3)=NOCH_3	
468	4-C(CH_3)=NOCH_3	m.p.: 105 - 107°C
469	3-C(CH_3)=NOC_2H_5	

No.	A_m	Data
470	4-C(CH_3)=NOC_2H_5	oil; IR (film): 3350, 2935, 1671, 1525, 1947, 979
471	3-C(CH_3)=NO-n-C_3H_7	
472	4-C(CH_3)=NO-n-C_3H_7	oil; IR (film): 3350, 2963, 2935, 1672, 1525, 1037, 979, 927
473	3-C(CH_3)=NO-i-C_3H_7	
474	4-C(CH_3)=NO-i-C_3H_7	
475	3-C(CH_3)=NO-n-C_4H_9	
476	4-C(CH_3)=NO-n-C_4H_9	oil; IR (film): 3350, 2957, 2935, 1673, 1525, 1038, 979
477	3-C(CH_3)=NO-s-C_4H_9	
478	4-C(CH_3)=NO-s-C_4H_9	
479	3-C(CH_3)=NO-i-C_4H_9	
480	4-C(CH_3)=NO-i-C_4H_9	oil; IR (film): 3350, 2958, 2935, 1673, 1525, 1039, 979, 927
481	3-C(CH_3)=NO-t-C_4H_9	
482	4-C(CH_3)=NO-t-C_4H_9	
483	3-C(CH_3)=NO-n-C_5H_{11}	
484	4-C(CH_3)=NO-n-C_5H_{11}	
485	3-C(CH_3)=NO-n-C_6H_{13}	
486	4-C(CH_3)=NO-n-C_6H_{13}	
487	2-Cl, 3-CH_3	
488	2-Cl, 4-CH_3	
489	2-Cl, 5-CH_3	
490	3-Cl, 4-CH_3	m.p.: 88 - 90°C
491	3-Cl, 5-CH_3	m.p.: 86 - 88°C
492	3-Cl, 2-CH_3	
493	2-CH_3, 4-Cl	
494	2-CH_3, 5-Cl	
495	3-CH_3, 4-Cl	m.p.: 88 - 90°C
496	3-Cl, 5-t-C_4H_9	

No.	A_m	Data
497	2-Cl, 4-t-C_4H_9	
498	3-Cl, 4-t-C_4H_9	
499	3-Cl, 4-Cyclohexyl	
500	2-F,4-CH_3	
501	3-F,4-CH_3	m.p.: 42 - 44°C
502	3-F,5-CH_3	
503	2-CH_3, 4-F	
504	3-CH_3, 4-F	m.p.: 75 - 78°C
505	2-F, 3-t-C_4H_9	
506	2-F, 4-t-C_4H_9	
507	3-F, 4-t-C_4H_9	
508	2-F, 4-OCH_3	m.p.: 65 - 6°C
509	2-Br, 4-CH_3	
510	3-Br, 4-CH_3	m.p.: 54 - 56°C
511	3-Br, 5-CH_3	
512	2-CH_3, 4-Br	
513	3-CH_3, 4-Br	m.p.: 97 - 99°C
514	2-Br, 3-t-C_4H_9	
515	2-Br, 4-t-C_4H_9	
516	3-Br, 4-t-C_4H_9	
517	2-Cl, 4-NO_2	
518	3-Cl, 4-NO_2	
519	3-Cl, 5-NO_2	
520	3-NO_2, 4-Cl	m.p.: 108 - 110°C
521	3-Cl, 4-(But-2-en-2-yl)	
522	3-Cl, 4-OCH_3	oil; IR (film): 3340, 2940, 1672, 1507, 1271, 1063, 1037
523	3-OCH_3, 4-Cl	
524	3-Cl, 4-OC_2H_5	oil; IR (film): 3345, 2940, 1671, 1505, 1270, 1060, 1038
525	3-Cl, 4-O-i-C_3H_7	oil; IR (film): 3345, 1280, 2940, 1671, 1525, 1502, 1269, 1037, 979
526	3-Cl, 4-O-t-C_4H_9	

No.	A_m	Data
527	2-Cl, 5-CF_3	
528	3-Cl, 4-CF_3	
529	3-Cl, 5-CF_3	
530	3-CF_3, 4-Cl	m.p.: 68 - 70°C
531	3-Cl, 4-CH_2Cl	
532	3-Cl, 4-OCF_3	
533	3-Cl, 5-OCF_3	
534	3-OCF_3, 4- Cl	
535	3-Cl, 4-OCH_2CF_3	
536	3-Cl, 4-OC_2F_5	
537	3-Cl, 4-OCF_2CHF_2	
538	3-NH_2, 4-Cl	
539	3-Cl, 4-O-C(=O)CH_3	
540	3-Cl, 4-S-t-C_4H_9	
541	3-Cl, 5-S-t-C_4H_9	
542	4-Cl, 3-S-t-C_4H_9	
543	3-F, 5-CF_3	m.p.: 76 - 78°C
544	2-CH_3, 4-CN	
545	3-NO_2, 4-CH_3	
546	2-CH_3, 4-OCH_3	
547	3-CH_3, 4-OCH_3	m.p.: 85 - 89°C
548	3-CH_3, 5-OCH_3	
549	3-OCH_3, 4-CH_3	
550	3-t-C_4H_9, 4-OCH_3	
551	3-t-C_4H_9, 5-OCH_3	
552	3-OCH_3, 4-t-C_4H_9	
553	2-CH_3, 4-O-i-C_3H_7	
554	3-CH_3, 4-O-i-C_3H_7	
555	3-O-i-C_3H_7, 4-CH_3	
556	3-t-C_4H_9, 4-O-i-C_3H_7	
557	3-t-C_4H_9, 5-O-i-C_3H_7	
558	3-O-i-C_3H_7, 4-t-C_4H_9	
559	3-t-C_4H_9, 4-O-t-C_4H_9	
560	2-CH_3, 4-CF_3	
561	2-CH_3, 5-CF_3	
562	3-CH_3, 4-CF_3	
563	3-CH_3, 5-CF_3	

No.	A_m	Data
564	3-CF_3, 4-CH_3	
565	2-CH_3, 4-OCF_3	
566	3-CH_3, 4-OCF_3	
567	3-CH_3, 5-OCF_3	
568	3-OCF_3, 4-CH_3	
569	3-CH_3, 4-OCH_2CF_3	
570	3-CH_3, 4-OC_2F_5	
571	3-CH_3, 4-OCF_2CHF_2	
572	3-CH_3, 4-S-t-C_4H_9	
573	3-CF_3, 4-OCH_3	
574	3-CF_3, 4-O-t-C_4H_9	
575	3-OCH_3, 4-CF_3	
576	2-CH_3, 4-$C(CH_3)=NOCH_3$	
577	3-CH_3, 4-$C(CH_3)=NOCH_3$	
578	3-Cl, 4-CH_3, 5-Cl	
579	3-Cl, 4-t-C_4H_9, 5-Cl	
580	3-Cl, 4-OCH_3, 5-Cl	
581	3-Cl, 4-O-t-C_4H_9, 5-Cl	
582	3-Cl, 4-NH_2, 5-Cl	
583	3-CO_2CH_3, 4-OCH_3	
584	3-NH_2, 4-OCH_3	
585	2-OH, 5-CH_3	m.p.: 128 - 131°C
586	2-OCH_3, 3-Cl, 5-Cl	m.p.: 65 - 67°C
587	3-Br, 4-OCH_3, 5-Br	
588	3-i-C_3H_7, 4-OCH_3, 5-i-C_3H_7	
589	3-NO_2, 4-OCH_3	m.p.: 118 - 120°C
590	3-(t-C_4H_9), 4-OCH_3, 5-(t-C_4H_9)	
591	3-Cl, 4-s-C_4H_9	m.p.: 58 - 60°C
592	3-Cl, 4-i-C_3H_7	m.p.: 91 - 92°C
593	3-Cl, 4-C_2H_5	
594	3-Cl, 4-OC_6H_5	
595	3-Cl, 4-$OCHF_2$	m.p.: 60 - 62°C

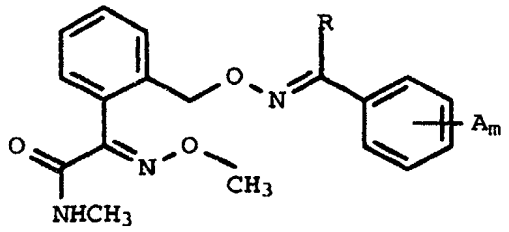
No.	A_m	Data
596	3-CF_3, 4-OC_6H_5	oil, IR (film): 3340, 2940, 1675, 1498, 1279, 1249, 1133, 1053, 1037
597	3-i-C_3H_7, 4-OCH_3	
598	3-CHF_2	oil, IR (film): 3340, 2940, 1670, 1528, 1208, 1036, 980
599	4-CHF_2	
600	3-Br, 4-OCH_3, 5-Cl	m.p.: 108 - 111°C
601	3-Br, 4-OC_6H_5, 5-Br	
602	3,5-$(CO_2CH_3)_2$	
603	3-OC_2H_5, - 4-Cl	
604	3-O-i-C_3H_7, 4-Cl	
605	3-O-t-C_4H_9, 4-Cl	
606	3-CH_3, 4-OC_2H_5	oil, IR (film): 3340, 2940, 1671, 1507, 1247, 1939
607	3-CH_3, 4-O-t-C_4H_9	
608	3-OC_2H_5, 4-CH_3	
609	3-O-t-C_4H_9, 4-CH_3	
610	3-Cl, 4-OCH_3, 5-CH_3	
611		m.p.: 92 - 95°C
612		oil, IR (film): 3350, 2932, 1671, 1525, 1037, 979, 777
613	3-Cl, 4-n-C_3H_7	oil; IR (film): 3349, 2937, 1671, 1506, 1271, 1037, 978

No.	A_m	Data
614	3-Cl, 4-O-Alkyl	oil; IR (film): 3340, 2930, 1672, 1504, 1270, 1037, 980
615	3-Cl, 4-O-n-C_4H_9	oil; IR (film): 3350, 2936, 1672, 1506, 1271, 1037, 979
616	3-Cl, 4-O-i-C_4H_9	oil; IR (film): 3345, 2960, 1670, 1506, 1271, 1059, 1037
617	3-Cl, 4-O-CH_2-CH=$CHCH_3$	oil; IR (film): 3345, 2937, 1673, 1504, 1269, 1037, 980
618	3-Cl, 4-O-CH_2-CH_2-CH_2-CH_2Cl	oil; IR (film): 3340, 2938, 1675, 1506, 1270, 1037
619	3-Cl, 4-O-n-C_5H_{11}	oil; IR (film): 3345, 2936, 1673, 1506, 1270, 1058, 1037, 980
620	3-Cl, 4-O-i-C_5H_{11}	oil; IR (film): 3340, 2955, 1672, 1506, 1269, 1060, 1037, 979
621	3-Cl, 4-O-CH_2-CH=$C(CH_3)_2$	oil; IR (film): 3345, 2940, 1674, 1504, 1269, 1037, 980
622	3-Cl, 4-O-neo-C_5H_{11}	oil; IR (film): 3345, 2956, 1673, 1507, 1259, 1061, 1038, 1014
623	3-Cl, 4-O-n-C_6H_{13}	oil; IR (film): 3417, 1681, 1267, 1034, 1026, 937
624	3-CH_3, 4-O-n-C_3H_7	oil; IR (film): 3340, 2937, 1671, 1506, 1038, 979

No.	A_m	Data
625	3-CH_3, 4-O-Alkyl	oil; IR (film): 3350, 2940, 1669, 1505, 1036, 980
626	3-CH_3, 4-O-n-C_4H_9	oil; IR (film): 3340, 2935, 1671, 1507, 12, 48, 1038, 979
627	3-CH_3, 4-O-CH_2-CH=$CHCH_3$	oil; IR (film): 3350, 2940, 1672, 1525, 1505, 1247, 1037, 979
628	3-CH_3, 4-O-n-C_5H_{11}	oil; IR (film): 3340, 2935, 1668, 1526, 1507, 1250, 1037, 980
629	3-CH_3, 4-O-i-C_5H_{11}	oil; IR (film): 3340, 2954, 1671, 1507, 1248, 1038, 980
630	3-CH_3, 4-O-CH_2-CH=$C(CH_3)_2$	oil; IR (film): 3350, 2935, 1672, 1505, 1037, 979
631	3-CH_3, 4-O-neo-C_5H_{11}	oil; IR (film): 3340, 2954, 1670, 1507, 1247, 1038
632	3-CH_3, 4-O-n-C_6H_{13}	oil; IR (film): 3345, 2934, 1672, 1506, 1247, 1038
633	4-O-CH_2-CH_2-CH_2-CH_2Nr	m.p.: 74 - 77°C
634	3,5-$(OCH_3)_2$, 4 OH	m.p.: 80 - 83°C
635	2,3,4,5,6-F_5	m.p.: 83 - 84°C
636	3,5-$(OH)_2$	m.p.: 68 - 70°C
637	4-O-i-C_5H_{11}	m.p.: 91 - 93°C
638	4-O-neo-C_5H_{11}	oil; IR (film): 3350, 2956, 1675, 1514, 1248, 1038, 1017

No.	A_m	Data
639	4-[O-CH_2-CH=C(CH_3)$_2$]	m.p.: 113 - 116°C
640	4-O-CH_2-CH_2-CH_2-CH_2Cl	m.p.: 77 - 80°C
641	4-O-CH_2.CH_2-CH_2CN	oil; IR (film): 3375, 2940, 2245, 1674, 1514, 1249, 1037
642	2-O-i-C_5H_{11}	oil; IR (film): 3350, 2955, 2935, 1672, 1525, 1222, 1038, 980
643	3-[O-CH_2-CH=C(CH_3)$_2$]	oil; IR (film): 3355, 2935, 1673, 1525, 1219, 1037, 980
644	3-O-CH_2-CH_2-CH_2-CH_2Cl	oil; IR (film): 3355, 2938, 1675, 1525, 1225, 1037, 979
645	4-NHC(=O)H	m.p.: 140 - 142°C
646	3-CH_3, 4 OH	m.p.: 159 - 161°C
647	3 CN	m.p.: 200 - 202°C
648	3-CH_3, 4-O-iC_3H_7	m.p.: 85 - 87°C
649	3-Cl, 4-OH	m.p.: 160 - 163°C
650	4-C(CH_3)$_2$CN	m.p.: 83 - 85°C
651	3-CF_3, 4 F	m.p.: 60 - 62°C

Table 2



No.	A_m	R	Data
1	H	C_2H_5	oil; IR (film): 3350, 2938, 1671, 1525, 1463, 1091, 1037, 979, 912, 770, 696
2	2-Cl	C_2H_5	m.p. 71 - 72°C
3	3-Cl	C_2H_5	m.p. 106 - 107°C
4	4-Cl	C_2H_5	
5	3,4-Cl_2	C_2H_5	m.p. 120 - 122°C
6	3,5-Cl_2	C_2H_5	
7	2,3,4-Cl_3	C_2H_5	
8	3,4,5-Cl_3	C_2H_5	
9	2-Br	C_2H_5	
10	3-Br	C_2H_5	
11	4-Br	C_2H_5	m.p. 107 - 108°C
12	2-NO_2	C_2H_5	
13	3-NO_2	C_2H_5	
14	4-NO_2	C_2H_5	
15	2-CH_3	C_2H_5	
16	3-CH_3	C_2H_5	
17	4-CH_3	C_2H_5	oil; IR (film): 3350, 2937, 1670, 1525, 1514, 1463, 1037, 979, 959, 911, 822
18	2-4-$(CH_3)_2$	C_2H_5	
19	3,4-$(CH_3)_2$	C_2H_5	

20	3,5-$(CH_3)_2$	C_2H_5	
21	3-i-C_3H_7	C_2H_5	
22	4-i-C_3H_7	C_2H_5	
23	3-t-C_4H_9	C_2H_5	
24	4-t-C_4H_9	C_2H_5	m.p. 94 - 95°C
25	3-CH_3, 4-t-C_4H_9	C_2H_5	
26	3-t-C_4H_9, 4-CH_3	C_2H_5	
27	3-OCH_3	C_2H_5	
28	4-OCH_3	C_2H_5	m.p. 96 - 97°C
29	3-O-i-C_3H_7	C_2H_5	
30	4-O-i-C_3H_7	C_2H_5	
31	3-O-t-C_4H_9	C_2H_5	
32	4-O-t-C_4H_9	C_2H_5	
33	3-CF_3	C_2H_5	
34	4-CF_3	C_2H_5	
35	3-OCF_3	C_2H_5	
36	4-OCF_3	C_2H_5	
37	3-S-t-C_4H_9	C_2H_5	
38	4-S-t-C_4H_9	C_2H_5	
39	3-$C(CH_3)=NOCH_3$	C_2H_5	
40	4-$C(CH_3)=NOCH_3$	C_2H_5	
41	H	n-C_3H_7	
42	2-Cl	n-C_3H_7	
43	3-Cl	n-C_3H_7	
44	4-Cl	n-C_3H_7	cf. Example 5
45	3,5-Cl_2	n-C_3H_7	
46	2,3,4-Cl_3	n-C_3H_7	
47	2-Br	n-C_3H_7	
48	3-Br	n-C_3H_7	
49	4-Br	n-C_3H_7	
50	2-CH_3	n-C_3H_7	
51	3-CH_3	n-C_3H_7	
52	4-CH_3	n-C_3H_7	
53	4-t-C_4H_9	n-C_3H_7	
54	4-CF_3	n-C_3H_7	
55	4-O-t-C_4H_9	n-C_3H_7	
56	4-OCF_3	n-C_3H_7	
57	H	i-C_3H_7	
58	2-Cl	i-C_3H_7	

59	3-Cl	i-C_3H_7	
60	4-Cl	i-C_3H_7	
61	3,5-Cl_2	i-C_3H_7	
62	2,3,4-Cl_3	i-C_3H_7	
63	2-Br	i-C_3H_7	
64	3-Br	i-C_3H_7	
65	4-Br	i-C_3H_7	
66	2-CH_3	i-C_3H_7	
67	3-CH_3	i-C_3H_7	
68	4-CH_3	i-C_3H_7	
69	4-t-C_4H_9	i-C_3H_7	
70	4-CF_3	i-C_3H_7	
71	4-O-t-C_4H_9	i-C_3H_7	
72	4-OCF_3	i-C_3H_7	
73	2-Cl	Cyclopropyl	
74	3-Cl	Cyclopropyl	
75	3,4-Cl_2	Cyclopropyl	
76	3,5-Cl_2	Cyclopropyl	
77	2,3,4-Cl_3	Cyclopropyl	
78	2-Br	Cyclopropyl	
79	3-Br	Cyclopropyl	
80	4-Br	Cyclopropyl	
81	2-CH_3	Cyclopropyl	
82	3-CH_3	Cyclopropyl	
83	4-CH_3	Cyclopropyl	
84	4-CF_3	Cyclopropyl	
85	4-O-t-C_4H_9	Cyclopropyl	
86	4-OCF_3	Cyclopropyl	
87	4F	C_2H_5	m.p. 94 - 95°C

The novel compounds are suitable as fungicides and insecticides.

The fungicidal compounds according to the invention, or agents containing them, may be applied for instance in the form of directly sprayable solutions, powders, suspensions (including high-percentage aqueous, oily or other suspensions), dispersions, emulsions, oil dispersions, pastes, dusts, broadcasting agents, or granules by spraying, atomizing, dusting, broadcasting or watering. The forms of application depend entirely on the purpose for which the agents are being used, but they must ensure as fine

a distribution of the active ingredients according to the invention as possible.

Normally, the plants are sprayed or dusted with the active ingredients or the seeds of the plants are treated with the active ingredients.

The formulations are produced in known manner, for example by extending the active ingredient with solvents and/or carriers, with or without the use of emulsifiers and dispersants; if water is used as solvent, it is also possible to employ other organic solvents as auxiliary solvents. Suitable auxiliaries for this purpose are solvents such as aromatics (e.g., xylene), chlorinated aromatics (e.g., chlorobenzenes), paraffins (e.g., crude oil fractions), alcohols (e.g., methanol, butanol), ketones (e.g., cyclohexanone), amines (e.g., ethanolamine, dimethylformamide), and water; carriers such as ground natural minerals (e.g., kaolins, aluminas, talc and chalk) and ground synthetic minerals (e.g., highly disperse silica and silicates); emulsifiers such as nonionic and anionic emulsifiers (e.g., polyoxyethylene fatty alcohol ethers, alkyl sulfonates and aryl sulfonates); and dispersants such as lignin-sulfite waste liquors and methylcellulose.

Examples of surfactants are: alkali metal, alkaline earth metal and ammonium salts of aromatic sulfonic acids, e.g., ligninsulfonic acid, phenolsulfonic acid, naphthalenesulfonic acid and dibutylnaphthalenesulfonic acid, and of fatty acids, alkyl and alkylaryl sulfonates, and alkyl, lauryl ether and fatty alcohol sulfates, and salts of sulfated hexadecanols, heptadecanols, and octadecanols, salts of fatty alcohol glycol ethers, condensation products of sulfonated naphthalene and naphthalene derivatives with formaldehyde, condensation products of naphthalene or naphthalenesulfonic acids with phenol and formaldehyde, polyoxyethylene octylphenol ethers, ethoxylated isooctylphenol, ethoxylated octylphenol and ethoxylated nonylphenol, alkylphenol polyglycol ethers, tributylphenyl polyglycol ethers, alkylaryl polyether alcohols, isotridecyl alcohol, fatty alcohol ethylene oxide condensates, ethoxylated castor oil, polyoxyethylene alkyl ethers, ethoxylated polyoxypropylene, lauryl alcohol polyglycol ether acetal, sorbitol esters, lignin-sulfite waste liquors and methyl cellulose.

Powders, dusts and broadcasting agents may be prepared by mixing or grinding the active ingredients with a solid carrier.

Granules, e.g., coated, impregnated or homogeneous granules, may be prepared by bonding the active ingredients to solid carriers. Examples of solid carriers are mineral earths such as silicic acids, silica gels, silicates, talc, kaolin, attapulgus clay, limestone, lime, chalk, bole, loess, clay, dolomite, diatomaceous earth, calcium sulfate, magnesium sulfate, magnesium oxide, ground plastics, fertilizers such as ammonium sulfate, ammonium phosphate, ammonium nitrate, and ureas, and vegetable products such as grain meals, bark meal, wood meal, and nutshell meal, cellulosic powders, etc.

Examples of formulations are given below.

I. A solution of 90 parts by weight of compound no. 5 from Table 1 (5/1) and 10 parts by weight of N-methyl-α-pyrrolidone, which is suitable for application in the form of very fine drops.

II. A mixture of 20 parts by weight of compound no. 6/1, 80 parts by weight of xylene, 10 parts by weight of the adduct of 8 to 10 moles of ethylene oxide and 1 mole of oleic acid-N-monoethanolamide, 5 parts by weight of the calcium salt of dodecylbenzenesulfonic acid, and 5 parts by weight of the adduct of 40 moles of ethylene oxide and 1 mole of castor oil. By finely dispersing the mixture in water, an aqueous dispersion is obtained.

III. An aqueous dispersion of 20 parts by weight of compound no. 8/1, 40 parts by weight of cyclohexanone, 30 parts by weight of isobutanol, 20 parts by weight of the adduct of 40 moles of ethylene oxide and 1 mole of castor oil.

IV. An aqueous dispersion of 20 parts by weight of compound no. 9/1, 25 parts by weight of cyclohexanol, 65 parts by weight of a mineral oil fraction having a boiling point between 210 and 280°C, and 10 parts by weight of the adduct of 40 moles of ethylene oxide and 1 mole of castor oil.

V. A hammer-milled mixture of 80 parts by weight of compound no. 10/1, 3 parts by weight of the sodium salt of diisobutylnaphthalene-α-sulfonic acid, 10 parts by weight of the sodium salt of a lignin-sulfonic acid obtained from a sulfite waste liquor, and 7 parts by weight of powdered silica gel. By finely dispersing the mixture in water, a spray liquor is obtained.

VI. An intimate mixture of 3 parts by weight of compound no. 11/1 and 97 parts by weight of particulate kaolin. The dust contains 3wt% of the active ingredient.

VII. An intimate mixture of 30 parts by weight of compound no. 16/1, 92 parts by weight of powdered silica gel and 8 parts by weight of paraffin oil sprayed onto the surface of this silica gel. This formulation of the active ingredient exhibits good adherence.

VIII. A stable aqueous dispersion of 40 parts by weight of compound no. 18/1, 10 parts of the sodium salt of a phenolsulfonic acid-urea-formaldehyde condensate, 2 parts of silica gel and 48 parts of water, which dispersion can be further diluted.

IX. A stable oily dispersion of 20 parts by weight of compound no. 19/1, 2 parts by weight of the calcium salt of dodecylbenzenesulfonic acid, 8 parts by weight of a fatty alcohol polyglycol ether, 2 parts by weight of the sodium salt of a phenolsulfonic acid-urea-formaldehyde condensate and 68 parts by weight of a paraffinic mineral oil.

The compounds are extremely effective on a broad spectrum of phytopathogenic fungi, in particular those from the class consisting of the Ascomycetes and Basidiomycetes. Some of them have a systemic action and can be used as foliar and soil fungicides.

The fungicidal compounds are of particular interest for controlling a large number of fungi in various crops or their seeds, especially wheat, rye, barley, oats, rice, Indian corn, lawns, cotton, soybeans, coffee, sugar cane, fruit and ornamentals in horticulture and viticulture, and in vegetables such as cucumbers, beans and cucurbits.

The compounds are applied by treating the fungi or the seeds, plants or materials to be protected against fungus attack, or the soil with a fungicidally effective amount of the active ingredients.

The active ingredients may be applied before or after infection of the materials, plants or seeds by the fungi.

Specifically, the compounds I are suitable for controlling the following plant diseases:

Erysiphe graminis in cereals,
Erysiphe cichoracearum and Sphaerotheca fuliginea in cucurbits,
Podosphaera leucotricha in apples,
Uncinula necator in vines,
Puccinia species in cereals,
Rhizoctonia solani in cotton,
Ustilago species in cereals and sugar cane,
Venturia inaequalis (scab) in apples,
Helminthosporium species in cereals,
Septoria nodorum in wheat,
Botrytis cinerea (gray mold) in strawberries and grapes,
Cercospora arachidicola in groundnuts,
Pseudocercosporella herpotrichoides in wheat and barley,
Pyricularia oryzae in rice,
Phytophthora infestans in potatoes and tomatoes,
Fusarium and Verticillium species in various plants,
Plasmopara viticola in grapes,
Alternaria species in fruit and vegetables.

The novel compounds may also be used for protecting materials (timber), for example against Paecilomyces variotii.

The fungicidal agents generally contain from 0.1 to 95, and preferably from 0.5 to 90, wt% of active ingredient.

The application rates depend on the type of effect desired, and vary from 0.02 to 3 kg/ha.

When the active ingredients are used for treating seed, rates of from 0.001 to 50, and preferably from 0.01 to 10, g per kg of seed are generally required.

When the agents according to the invention are used as fungicides, they may be applied together with other active ingredients, for example herbicides, insecticides, growth regulators, other fungicides and fertilizers.

When mixed with other fungicides, the spectrum of fungicidal action is frequently increased.

Use examples

For the following use examples the comparative substances employed were methyl 2-methoximino-2-[2'-(1''-(1'''-naphthyl)-1''-methyl)-iminooxymethyl]-phenylacetate (A), methyl 2-methoximino-2-[2'-(1''-(2'''-naphthyl)-1''-methyl)-iminooxymethyl]-phenylacetate (B) and methyl 2-methoximino-2-[2'-(1''-(1'''-

phenyl)-1''-methyl)-iminooxymethyl]-phenylacetate (C) - all known from EP 463 488.

Use Example 1

Action on Fusarium culmorum in wheat

Leaves of pot-grown wheat seedlings of the "Kanzler" variety were sprayed to runoff with aqueous liquors containing (dry basis) 80% of active ingredient and 20% of emulsifier. The following day they were inoculated with a spore suspension of Fusarium culmorum and then placed in a climatic cabinet of high humidity (> 90%) at 22-24°C. The extent of fungus spread was assessed visually after 6 days.

Active ingredient	Leaf attack in % after application of aqueous formulations containing 500 ppm of active ingredient
Table 1, No. 1	0
Table 1, No. 2	5
Comparative product A	70
Comparative product B	25
Untreated	70

Use Example 2
Action on Plasmopara viticola

Leaves of potted vines of the Müller-Thurgau variety were sprayed with aqueous suspensions containing (dry basis) 80% of active ingredient and 20% of emulsifier. To assess the duration of action, the plants were set up, after the sprayed-on layer had dried, for 8 days in the greenhouse. Then the leaves were infected with a zoospore suspension of Plasmopara viticola. The plants were first placed for 48 hours in a water vapor-saturated chamber at 24°C and then in a greenhouse for 5 days at from 20 to 30°C. To accelerate and intensify the sporangiophore discharge, the plants were then again placed in the moist chamber for 16 hours. The extent of fungus attack was then assessed on the undersides of the leaves.

Active ingredient	Leaf attack in % after application of aqueous formulations containing 16 ppm of active ingredient
Table 1, No. 5	5
Table 1, No. 19	0
Table 1, No. 63	5
Table 1, No. 74	15
Table 1, No. 118	5
Table 1, No. 228	5
Table 1, No. 282	0
Table 1, No. 283	5
Comparative product C	25
Untreated	70

Use example 3
Action on wheat brown rust

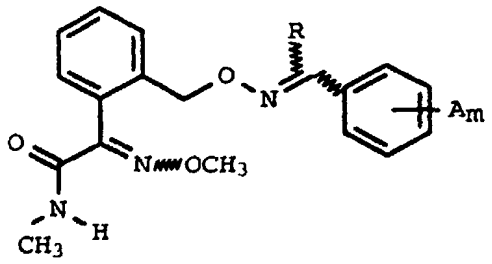
Leaves of pot-grown wheat seedlings of the "Frühgold" variety were dusted with spores of brown rust (Puccinia recondita). The pots were then placed for 24 hours at 20 to 22°C in a high-humidity (90 - 95%) chamber. During this period the spores germinated and the germ tubes penetrated the leaf tissue. The infected plants were then sprayed to runoff with aqueous liquors containing (dry basis) 80% of active ingredient and 20% of emulsifier. After the sprayed-on layer had dried, the plants were set up in the greenhouse at 20 to 22°C and a relative humidity of 65 to 70%. The extent of rust fungus spread on the leaves was assessed after 8 days.

Active ingredient	Leaf attack in % after application of aqueous formulations containing 63 ppm of active ingredient
Table 1, No. 1	0
Table 1, No. 2	5
Table 1, No. 19	15
Table 1, No. 63	0
Table 1, No. 118	5
Table 1, No. 282	0
Table 1, No. 283	0
Comparative product A	35
Comparative product B	25
Comparative product C	65
Untreated	65

The results obtained in these comparative experiments show that the active ingredients from Table 1, Nos. 1, 2, 5, 19, 63, 74, 118, 228, 282 and 283, when applied in the stated concentrations in spray liquors, have a better fungicidal action than the prior art comparative active ingredients A, B and C.

We claim:

1. An N-methylamide of the formula I

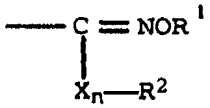


I,

where

R is C_1-C_3-alkyl or cyclopropyl,

A is identical or different and is hydrogen, halogen, cyano, nitro, C_1-C_6-alkyl, C_3-C_6-cycloalkyl, OR^1, C_3-C_6-cycloalkyloxy, C_1-C_6-haloalkyl, C_1-C_6-haloalkyloxy, C_2-C_6-alkenyl, C_2-C_6-alkenyloxy, C_2-C_6-alkynyl, C_1-C_6-alkoxy-C_1-C_6-alkyl, cyano-C_1-C_6-alkyl, nitro-C_1-C_6-alkyl, phenyl, phenoxy, $C(O)R^1$, CO_2R^1, $C(O)NR^1R^2$, $C(S)NR^1R^2$, NR^1R^2, $NR^1C(O)R^2$, $NR^1CO_2R^2$, $OC(O)R^1$, SR^1, $S(O)R^1$, $S(O)_2R^1$, $-C(R^1)=NR^2$, $-N=CR^1R^2$,



where R^1, R^2 and R^3 are identical or different and each is hydrogen or C_1-C_6-alkyl and X is S, O or NR^3 and n is 0 or 1 and two of the groups A_m in adjacent positions may be $-CH=CH-CH=CH-$ and

m is 1, 2 or 3, with the exception of compounds of the formula I in which

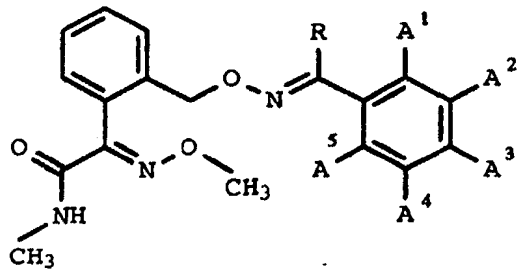
R is methyl and

A_m is 2-chloro, 3-chloro, 4-chloro, 3,5-dichloro, 2,3,4-trichloro, 2-methyl, 4-methyl, 3-bromo, 4-nitro or hydrogen, and with the exception of compounds of the formula I in which

R is cyclopropyl and

A_m is hydrogen, 4-chloro, 4-tert.-butyl or 4-methoxy.

2. An N-methylamide of the formula Ia



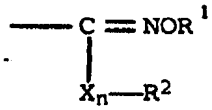
Ia,

where

R is methyl, ethyl, n-propyl or isopropyl,

A^1, A^2, A^3, A^4 and A^5 are identical or different and each is hydrogen, halogen, cyano, nitro, C_1-C_6-alkyl, C_3-C_6-cycloalkyl, OR^1, C_3-C_6-cycloalkyloxy, C_1-C_6-haloalkyl, C_1-C_6-haloalkyloxy, C_2-C_6-alkenyl, C_2-C_6-alkenyloxy, C_2-C_6-alkynyl, C_1-C_6-alkoxy-C_1-C_6-alkyl, cyano-C_1-C_6-alkyl, nitro-C_1-C_6-alkyl, phenyl, phenoxy, $C(O)R^1$, CO_2R^1, $C(O)NR^1R^2$, $C(S)NR^1R^2$, NR^1R^2, $NR^1C(O)R^2$, $NR^1CO_2R^2$, $OC(O)R^1$, SR^1, $S(O)R^1$, $S(O)_2R^1$,

$-C(R^1)=NR^2$, $-N=CR^1R^2$,



where R^1, R^2 and R^3 are identical or different and each is hydrogen or C_1-C_6-alkyl and X is S, O or NR^3 and n is 0 or 1, and two of the groups A^1 to A^5 in adjacent positions may be $-CH=CH-CH=CH-$,

with the proviso that

a) two to four of the substituents A^1 to A^5 are hydrogen,

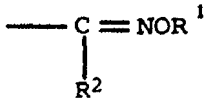
b) two to three of the substituents A^1 to A^5 are hydrogen when one of the substituents A^1 to A^5 is Cl, Br, NO_2 or CH_3,

c) one of the halogen atoms is F or Br when A^2+A^4 or $A^1+A^2+A^3$ are at the same time halogen.

3. An N-methylamide of the formula Ia as set forth in claim 2, where

R is methyl, ethyl, n-propyl or isopropyl,

A^1, A^2, A^3, A^4 and A^5 are identical or different and each is hydrogen, halogen, nitro, cyano, C_1-C_6-alkyl, C_1-C_6-alkoxy, C_1-C_6-alkoxyalkyl, C_1-C_6-haloalkyl, C_1-C_6-haloalkoxy or



where R^1 and R^2 are identical of different and each is hydrogen or C_1-C_6-alkyl, with the proviso that

a) two to four of the substituents A^1 to A^5 are hydrogen,

b) two to three of the substituents A^1 to A^5 are hydrogen when one of the substituents A^1 to A^5 is Cl, Br, NO_2 or CH_3,

c) one of the halogen atoms is F or Br when A^2+A^4 or $A^1+A^2+A^3$ are at the same time halogen.

4. An N-methylamide of the formula Ia as set forth in claim 2, where R is methyl, A^3, A^4 and A^5 are hydrogen and A^1 and A^2 are -CH=CH-CH=CH-.

5. An N-methylamide of the formula Ia as set forth in claim 2, where R is methyl, A^1, A^4 and A^5 are hydrogen and A^2 and A^3 are -CH=CH-CH=CH-.

6. A fungicide containing an inert carrier and a fungicidally effective amount of an N-methylamide of the formula I as set forth in claim 1.

7. A process for combating fungi, wherein the fungi or the materials, plants, seed or the soil threatened by fungus attack are treated with a fungicidally effective amount of a compound of the formula I as set forth in claim 1.